

(19) World Intellectual Property
Organization
International Bureau



(43) International Publication Date
2 December 2004 (02.12.2004)

PCT

(10) International Publication Number
WO 2004/104127 A2

(51) International Patent Classification⁷: **C09J 123/08**,
C08L 23/08

(21) International Application Number:
PCT/US2004/015697

(22) International Filing Date: 19 May 2004 (19.05.2004)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
60/471,318 19 May 2003 (19.05.2003) US
10/666,488 19 September 2003 (19.09.2003) US

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(81) Designated States (*unless otherwise indicated, for every
kind of national protection available*): AE, AG, AL, AM,

AT, AU, AZ, BA, BB, BG, BR, BW, BY, BZ, CA, CH, CN,
CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, EG, ES, FI,
GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE,
KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD,
MG, MK, MN, MW, MX, MZ, NA, NI, NO, NZ, OM, PG,
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TN, TR, TT, TZ, UA, UG, UZ, VC, VN, YU, ZA, ZM, ZW.

(84) Designated States (*unless otherwise indicated, for every
kind of regional protection available*): ARIPO (BW, GH,
GM, KE, LS, MW, MZ, NA, SD, SL, SZ, TZ, UG, ZM,
ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM),
European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI,
FR, GB, GR, HU, IE, IT, LU, MC, NL, PL, PT, RO, SE, SI,
SK, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ,
GW, ML, MR, NE, SN, TD, TG).

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earlier application (Rule 4.17(iii)) for all designations*

Published:

- *without international search report and to be republished
upon receipt of that report*

*For two-letter codes and other abbreviations, refer to the "Guid-
ance Notes on Codes and Abbreviations" appearing at the begin-
ning of each regular issue of the PCT Gazette.*

(54) Title: NOVEL MULTIFUNCTIONAL POLYMER FOR USE IN HOT MELT ADHESIVE APPLICATIONS

(57) Abstract: Adhesives comprising one or more tackifiers and an ethylene/alpha-olefin interpolymer produced using either a single metallocene or dual metallocene catalyst system were formulated, and have adhesive characteristics over a broad temperature range. The inventive hot melt adhesive compositions ("HMAs") have properties comparable to those of commercially available, three component HMAs comprising ethylene vinyl acetate polymers, tackifier and wax. HMA embodiments include those wherein the ethylene/alpha-olefin interpolymers comprise ethylene and either 1-octene or propylene as copolymers.



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**NOVEL MULTIFUNCTIONAL POLYMER FOR USE IN HOT MELT
ADHESIVE APPLICATIONS.**

CROSS REFERENCES TO RELATED APPLICATIONS.

This application claims the benefit of U.S. Provisional Patent Application, Serial No. 60/471,318, filed 19 May 2003, and U.S. Non-Provisional Patent Application, Serial No. 10/666,488 filed 19 September 2003, the contents of which are hereby incorporated by reference herein.

FIELD OF THE INVENTION.

The present invention is a novel hot melt adhesive composition consisting essentially of a selected ethylene/alpha-olefin interpolpolymer, and optionally one or more tackifiers. Unlike conventional hot melt adhesives, which consist of three separate components, a polymer, a wax, and a tackifier, the hot melt adhesive of the present invention employs an ethylene/ α -olefin interpolpolymer. This interpolpolymer is carefully selected as to its composition and properties, so as to function, in an adhesive composition, as both the polymer and the wax. Thus, the adhesive can comprise either a single component for low tack applications, or a simple two component adhesive for applications that require the addition of a tackifier.

BACKGROUND OF THE INVENTION

Hot melt adhesives ("HMA's") are ubiquitous in many areas of commerce including consumer and industrial packaging where a bond is required between a substrate and a second item. They are routinely used in the manufacture of corrugated cartons, boxes and the like. They are also used in diverse areas, such as bookbinding; sealing the ends of paper bags; furniture manufacturing; manufacture of particleboard, linerboard, various other paper goods, and for adhering other articles, such as glass, metals and various plastics, including attaching paper labels to plastic containers. Additional uses of hot-melt adhesives also include, carpet seam sealing tape, lamination, product assembly, non-woven construction, and potting and encapsulation compounds.

1 Because of these diverse applications, hot melt adhesives may be required to
2 maintain a strong bond over a wide range of temperature conditions. For example, in the
3 manufacture of corrugated cartons used for shipping refrigerated or frozen foods, or
4 foods packed in ice, hot melt adhesives are generally selected because of their ability to
5 maintain a strong bond under low temperature conditions. However in other applications
6 the hot melt adhesive may have to maintain a strong bond to the substrate under extremes
7 of stress and shock in handling, and high humidity.

8 Unlike other adhesives, which are often applied as a solution in a solvent, HMA's
9 are generally solids, and, in commercial applications, are typically applied to substrates in
10 their molten state at temperatures of about 350 degrees F. As the molten adhesive cools
11 and solidifies, a bond is formed between the substrate and the second item. Various
12 techniques can be used to apply hot melt adhesives to a substrate including roll coaters,
13 knife coaters and spray devices.

14 Two other important factors in hot melt adhesive performance are the so-called
15 "set time" and "open time" of the adhesive. The "open time" of a hot melt adhesive is the
16 time it takes to solidify to a point where it can no longer bond with the intended article.
17 The "set time" of a hot melt adhesive is the time required for the adhesive to cool to the
18 point where it has enough strength to form a bond. Set speed is an important parameter
19 for applications such as high speed packaging lines, where bonding needs to occur
20 rapidly to avoid poorly sealed or unsealed boxes.

21 Most hot melt adhesives are mixtures of three components: a wax, a tackifying
22 agent and a polymeric resin. Although each component is generally present in roughly
23 equal proportions in an HMA formulation, their relative ratio is often "fine tuned" for a
24 particular application's need.

25 The polymer component provides the strength to the adhesive bond. The tackifier
26 provides tack to the adhesive by improving wetting, which serves to secure the items to
27 be bonded while the adhesive sets, and reduces the viscosity of the system making the
28 adhesive easier to apply to the substrate. The wax shortens the open time and also
29 reduces the viscosity of the system. In general, the percent wax is minimized and added
30 in quantities sufficient to achieve the desired viscosity and set speeds.

1 A number of hot melt adhesive formulations utilize a vinyl acetate ("VA")
2 polymer as the polymer component and the formulations are varied according to the vinyl
3 acetate content of the polymer. Low vinyl acetate content polymers are preferred due to
4 their lower cost, and as they are relatively non polar, they can be formulated with other
5 relatively non-polar tackifiers and waxes to yield compatible formulations. Higher vinyl
6 acetate content polymer resins (with greater than about 18% vinyl acetate content) when
7 used in hot melt adhesive formulations result in a stronger ionic bond to polar substrates
8 such as paper, thereby creating a stronger adhesive. However, the use of higher vinyl
9 acetate content polymers requires formulating with more polar waxes and tackifiers to
10 maintain formulation compatibility. More polar waxes, such as Fischer-Tropsch ("FT")
11 waxes are generally more expensive than paraffin wax and the selection and supply of
12 these more polar waxes is limited. They are difficult to obtain domestically and are thus
13 potentially subject to supply interruptions caused by world events.

14 In addition to bonding requirements, HMA's require performance in other areas
15 such as thermal and oxidative stability. Hot melt adhesives are applied in a molten state;
16 consequently many applications involve prolonged exposure to high temperatures. Good
17 thermal and oxidative stability means that the HMA will not darken nor produce a char or
18 skin or gel, nor will it exhibit a substantial viscosity change over time. Such charring,
19 skinning, gel formation and/or viscosity changes also increase the propensity of the
20 formulation to cause plugged lines and nozzles while in use, as in industrial applications.
21 The introduction of any wax into an HMA formulation, and especially the more polar
22 waxes, tends to lower the formulation's thermal and oxidative stability.

23 Hot melt adhesives comprised of ethylene polymers other than those
24 incorporating vinyl acetate have also been disclosed in the prior art. For instance, U.S.
25 Patent No. 5,021,257, issued on June 4th, 1991, to Foster et al., discloses a hot- melt
26 adhesive composition having a viscosity of about 3,000 to about 25,000 centipoise at
27 135°C, and a Ring and Ball softening point of about 90°C to about 125°C, said adhesive
28 composition comprising a blend of at least one substantially amorphous
29 propylene/hexene copolymer, at least one tackifier, and at least one substantially
30 crystalline, low viscosity hydrocarbon wax.

1 U.S. Pat. No. 5,530,054, issued Jun. 25, 1996 to Tse et al., claims a hot melt
2 adhesive composition consisting essentially of: (a) 30 percent to 70 percent by weight of
3 a copolymer of ethylene and about 6 percent to about 30 percent by weight of a C₃ to C₂₀
4 α -olefin produced in the presence of a catalyst composition comprising a metallocene
5 and an alumoxane and having an M_w of from about 20,000 to about 100,000; and (b) a
6 hydrocarbon tackifier which is selected from a recited list.

7 U.S. Pat. No. 5,548,014, issued Aug. 20, 1996 to Tse et al., claims a hot melt
8 adhesive composition comprising a blend of ethylene/alpha-olefin copolymers wherein
9 the first copolymer has a M_w from about 20,000 to about 39,000 and the second
10 copolymer has a M_w from about 40,000 to about 100,000. Each of the hot melt adhesives
11 exemplified comprises a blend of copolymers, with at least one of the copolymers having
12 a polydispersity greater than 2.5. Furthermore, the lowest density copolymer exemplified
13 has a specific gravity of 0.894 g/cm³.

14 U.S. Patent No. 6,107,430, issued on August 22, 1991, to Dubois et al., discloses
15 hot melt adhesives comprising at least one homogeneous linear or substantially linear
16 interpolymers of ethylene with at least one C₂ - C₂₀ α -olefin interpolymers having a density
17 from 0.850 to 0.895 g/cm³, optionally at least one tackifying resin; and optionally at least
18 one wax, wherein the hot melt adhesive has a viscosity of less than about 5000 cP at
19 150°C.

20 Also, EP 0 886 656 B1, published on September 19, 2001, to Simmons et al.,
21 discloses hot melt adhesives comprising from 5 to 95 weight percent at least one
22 homogeneous linear or substantially linear interpolymers of ethylene with at least one α -
23 olefin interpolymers having a polydispersity index, M_w/M_n, of from 1.5 to 2.5, and a
24 density from 0.850 to 0.885 g/cm³, from 5 to 95 weight percent of at least one tackifying
25 resin; and optionally at least one wax.

26 Tse, in Application of Adhesion Model for Developing Hot Melt Adhesives
27 Bonded to Polyolefin Surfaces, Journal of Adhesion, Vol. 48, Issue 1-4, pp. 149-167,
28 1995, notes that compared with hot melt adhesives based on ethylene-vinyl acetate
29 copolymer, hot melt adhesives based on homogeneous linear ethylene/.alpha.-olefin
30 interpolymers show higher viscosity and inferior tensile strength, but better bond strength
31 to polyolefin surfaces, higher strain at break and lower yield stress.

1 Hot melt adhesives comprising these polymers can be made which match the
2 strength performance of the vinyl-acetate containing HMA formulations, but their ability
3 to be formulated with non polar tackifiers render the resulting hot melt formulation more
4 thermally stable than vinyl acetate containing hot melt adhesives.

5 However, neither the prior art involving vinyl acetate-based adhesives nor the
6 prior art involving non-vinyl acetate containing polymer-based adhesives anticipates the
7 present invention whereby a single synthetic polymer can be created that can substitute
8 for both the wax and polymer components of a hot melt adhesive formulation.

9 Such a low cost hot melt adhesive formulation, which is composed from a single
10 component (other than a tackifier) and which can be shipped and unloaded in molten
11 form would be highly advantageous. It would also be highly advantageous to have an
12 HMA formulation, which can be prepared with a minimum of mixing steps, thus
13 minimizing the cost and variability of the formulation. It would also be highly
14 advantageous to have an HMA formulation which is able to match the adhesion
15 performance of HMA's comprising high VA containing ethylene-vinyl acetate ("EVA")
16 polymers but without the requirement of incorporating expensive petroleum waxes that
17 are primarily imported and/or derived from imported oil based feedstocks. It would also
18 be highly advantageous if such hot melt adhesive formulations were able to exhibit the
19 strength and adhesion characteristics of the EVA-containing formulations while having
20 good thermal and oxidative stability.

21 The HMAs of the present invention comprise a single polymer component, which
22 functions as both the polymer and the wax, and which can readily be shipped and/or
23 unloaded in a molten state. For low tack applications, no additional components are
24 required whereas for higher tack applications one or more tackifiers can be added. Thus
25 the HMA compositions of the present invention require a minimum of mixing steps, each
26 of which introduce both additional cost and variability to the final HMA formulation.
27 The HMA compositions of the present invention can function without the requirement of
28 an expensive polar wax in the formulation.

29 The HMA compositions of the present invention also exhibit adhesion and
30 strength properties that are comparable to those of commercially available EVA-
31 containing hot melt adhesives, and also exhibit good thermal and oxidative stability. In

1 addition, the HMA compositions of the present invention provide a composition that,
2 when applied to consumer packaging that is subsequently recycled, can be recycled more
3 easily than conventional hot melt adhesives due to elimination of the wax component
4 and/or the reduced amounts of tackifier.

5 Finally, the formulations of the present invention provide a composition for use in
6 hot melt adhesives and for paper coating that has properties that are generally regarded as
7 safe by the Food and Drug Administration.

1
2 BRIEF SUMMARY OF THE INVENTION

3 The present invention comprises hot melt adhesive compositions having one or more
4 tackifiers and an ethylene/ α -olefin interpolymers. These ethylene/ α -olefin polymers were
5 synthesized using either a single or a dual metallocene catalyst polymerization process.
6

7 An embodiment of the present invention is a hot melt adhesive composition
8 consisting essentially of:

- 9 A) from about 40 to 100 percent by weight (based on the final weight of
10 the hot melt adhesive composition) of a homogenous ethylene/ α -olefin interpolymers; and
11 B) from 0 to about 60 percent by weight (based on the final weight of the
12 hot melt adhesive composition) of one or more tackifiers.
13

14 In another embodiment, the present invention is a hot melt adhesive composition
15 wherein:

16 A) the homogenous ethylene/ α -olefin interpolymers is present in an
17 amount of from about 60 to about 85 percent by weight (based on the final weight of the
18 hot melt adhesive composition) and the homogenous ethylene/ α -olefin interpolymers is
19 characterized by having:

- 20 i) a density of from about 0.880 to about 0.930 g/cm³;
21 ii) a number average molecular weight (Mn) of from about 1,000
22 to about 9,000; and
23 iii) a Brookfield Viscosity (measured at 300°F) of from about 500
24 to about 7,000 cP and

25 B) the one or more tackifiers is present in an amount of from about 15 to
26 about 40 percent by weight (based on the final weight of the hot melt
27 adhesive composition); and wherein

28 C) the hot melt adhesive composition is characterized by having:

- 29 i) a Brookfield Viscosity (measured at 350°F) of from about 400 to
30 about 2,000 cP;

1 ii) a Peel Adhesion Failure Temperature ("PAFT") of greater than
2 or equal to 110°F; and
3 iii) a Shear Adhesion Failure Temperature ("SAFT") of greater
4 than or equal to 140°F.

5
6 In a third embodiment, the present invention is a hot melt adhesive composition
7 wherein:

8 A) the homogenous ethylene/ α -olefin interpolymer is characterized by
9 having:

10 i) a density of from about 0.893 to about 0.930 g/cm³;
11 ii) a number average molecular weight (Mn) of from about 1,000
12 to about 6,000; and
13 iii) a Brookfield Viscosity (measured at 300°F) of from about
14 1,500 to about 5,000 cP; and

15 B) the hot melt adhesive composition is characterized by:

16 i) having a Brookfield Viscosity (measured at 350°F) of from
17 about 400 to about 1,400 cP;
18 ii) having a Peel Adhesion Failure Temperature ("PAFT") of
19 greater than or equal to 90°F;
20 iii) having a Shear Adhesion Failure Temperature ("SAFT") of
21 greater than or equal to 200°F.; and
22 iv) exhibits 100% paper tear at 120°F.

23
24 The adhesive characteristics of the inventive hot melt adhesive compositions
25 ("HMAs") were tested and were comparables to commercially available, three
26 component hot-melt adhesive formulations which comprise a polymer, a wax and a
27 tackifier.

1 DETAILED DESCRIPTION OF THE INVENTION

2 Unless indicated otherwise, the following testing procedures and
3 definitions are to be employed:

4 Melt index (I_2), is measured in accordance with ASTM D-1238, condition
5 190°C/2.16 kg (formally known as "Condition (E)").

6 Molecular weight is determined using gel permeation chromatography
7 (GPC) on a Waters 150°C high temperature chromatographic unit equipped with
8 three mixed porosity columns (Polymer Laboratories 103, 104, 105, and 106),
9 operating at a system temperature of 140°C. The solvent is 1,2,4-trichlorobenzene,
10 from which 0.3 percent by weight solutions of the samples are prepared for
11 injection. The flow rate is 1.0 mL/min. and the injection size is 100 microliters.

12 The molecular weight determination is deduced by using narrow molecular
13 weight distribution polystyrene standards (from Polymer Laboratories) in
14 conjunction with their elution volumes. The equivalent polyethylene molecular
15 weights are determined by using appropriate Mark-Houwink coefficients for
16 polyethylene and polystyrene (as described by Williams and Word in Journal of
17 Polymer Science, Polymer Letters, Vol. 6, (621) 1968) to derive the following
18 equation:

$$19 \quad M_{\text{polyethylene}} = a * (M_{\text{polystyrene}})^b.$$

20 In this equation, $a = 0.4316$ and $b = 1.0$. Weight average molecular weight, M_w , is
21 calculated in the usual manner according to the following formula: $M_w = \sum w_i * M_i$,
22 where w_i and M_i are the weight fraction and molecular weight, respectively, of
23 the i th fraction eluting from the GPC column.

24 Melt viscosity is determined in accordance with the following procedure
25 using a Brookfield Laboratories DVII+ Viscometer in disposable aluminum
26 sample chambers. The spindle used is a SC-31 hot-melt spindle, suitable for
27 measuring viscosities in the range of from 10 to 100,000 centipoise. A cutting

blade is employed to cut samples into pieces small enough to fit into the 1 inch wide, 5 inches long sample chamber. The sample is placed in the chamber, which is in turn inserted into a Brookfield Thermosel and locked into place with bent needle-nose pliers. The sample chamber has a notch on the bottom that fits the bottom of the Brookfield Thermosel to ensure that the chamber is not allowed to turn when the spindle is inserted and spinning. The sample is heated to the desired temperature, such as 300°F or 350°F, with additional sample being added until the melted sample is about 1 inch below the top of the sample chamber. The viscometer apparatus is lowered and the spindle submerged into the sample chamber. Lowering is continued until brackets on the viscometer align on the Thermosel. The viscometer is turned on, and set to a shear rate which leads to a torque reading in the range of 30 to 60 percent. Readings are taken every minute for about 15 minutes, or until the values stabilize, which final reading is recorded.

Percent crystallinity is determined by differential scanning calorimetry using a TA-Q1000. The percent crystallinity may be calculated with the equation :

$$\text{percent C} = (A/292 \text{ J/g}) \times 100,$$

wherein percent C represents the percent crystallinity and A represents the heat of fusion of the ethylene in Joules per gram (J/g).

Density is measured in accordance with ASTM D-792. The samples are annealed at ambient conditions for 24 hours before the measurement is taken.

Comonomer and monomer incorporation was determined using nuclear magnetic resonance (NMR) spectroscopy. ¹³C NMR analysis was used to determine ethylene content and comonomer content using the following procedures:.

¹³C NMR analysis

The samples were prepared by adding approximately 3g of a 50/50 mixture of tetrachloroethane-d₂/orthodichlorobenzene that is 0.025M in chromium acetylacetonate (relaxation agent) to 0.4g sample of polymer in a 10mm NMR tube. The samples were

1 dissolved and homogenized by heating the tube and its contents to 150°C. The data was
2 collected using a Varian Unity Plus 400MHz spectrometer, corresponding to a ^{13}C
3 resonance frequency of 100.4 MHz. Acquisition parameters were selected to ensure
4 quantitative ^{13}C data acquisition in the presence of the relaxation agent. The data was
5 acquired using gated ^1H decoupling, 4000 transients per data file, a 6sec pulse repetition
6 delay, spectral width of 24,200Hz and a file size of 32K data points, with the probe head
7 heated to 130°C.

8
9 The term "interpolymer" is used herein to indicate a copolymer, or a terpolymer,
10 or the like. That is, at least one other comonomer is polymerized with ethylene to make
11 the interpolymer.

12 The term "narrow composition distribution" used herein describes the comonomer
13 distribution for homogeneous interpolymers. The narrow composition distribution
14 homogeneous interpolymers can also be characterized by their SCBDI (short chain
15 branch distribution index) or CDBI (composition distribution branch index). The SCBDI
16 or CDBI is defined as the weight percent of the polymer molecules having a comonomer
17 content within 50 percent of the median total molar comonomer content.

18 The CDBI of a polymer is readily calculated from data obtained from techniques
19 known in the art, such as, for example, temperature rising elution fractionation
20 (abbreviated herein as "TREF") as described, for example, in Wild et al, Journal Of
21 Polymer Science, Poly. Phys. Ed., Vol. 20, p. 441 (1982), or in U.S. Patent No.
22 5,548,014, the disclosures of which are incorporated herein by reference. Thus, the
23 following procedure for calculating CDBI can be used:

- 24
- 25 (1) Generate a normalized, cumulative distribution plot of copolymer
26 concentration versus elution temperature, obtained from the TREF.
 - 27 (2) Determine the elution temperature at which 50 weight percent of the
28 dissolved copolymer has eluted.
 - 29 (3) Determine the molar comonomer content within the copolymer fraction
30 eluting at that median elution temperature.

- (4) Calculate limiting mole fraction values of 0.5 times and 1.5 times the molar comonomer content within the copolymer fraction eluting at that median temperature.
- (5) Determine limiting elution temperature values associated with those limiting mole fraction values.
- (6) Partially integrate that portion of the cumulative elution temperature distribution between those limiting elution temperature values.
- (7) Express the result of that partial integration, CDBI, as a percentage of the original, normalized, cumulative distribution plot.

a) Homogeneous Ethylene/ α -Olefin Interpolymer Component

By the term "homogeneous interpolymer" is used herein to indicate a linear or substantially linear ethylene interpolymer prepared using a constrained geometry or single site metallocene catalyst. By the term homogenous, it is meant that any comonomer is randomly distributed within a given interpolymer molecule and substantially all of the interpolymer molecules have the same ethylene/comonomer ratio within that interpolymer. The melting peak of homogeneous linear and substantially linear ethylene polymers, as determined by differential scanning calorimetry (DSC), will broaden as the density decreases and/or as the number average molecular weight decreases.

The homogeneous linear or substantially linear ethylene polymers can be characterized as having a narrow molecular weight distribution (M_w/M_n). For the linear and substantially linear ethylene polymers, the M_w/M_n is preferably from 1.5 to 2.5, preferably from 1.8 to 2.2. However, certain interpolymers of the present invention may have much larger values of M_w/M_n , and still exhibit excellent adhesive properties.

It is important to note that the ethylene polymers useful in the invention differ from low density polyethylene prepared in a high pressure process. In one regard, whereas low density polyethylene is an ethylene homopolymer having a density of from 0.900 to 0.935 g/cm³, the ethylene polymers useful in the invention require the presence of a comonomer to reduce the density to less than 0.935 g/cm³.

1 Substantially linear ethylene polymers are homogeneous polymers having long
2 chain branching. The long chain branches have the same comonomer distribution as the
3 polymer backbone and can be as long as about the same length as the length of the
4 polymer backbone. When a substantially linear ethylene polymer is employed in the
5 practice of the invention, such polymer may be characterized as having a polymer
6 backbone substituted with from 0.1 to 3 long chain branches per 1000 carbons.

7 For quantitative methods for determination, see, for instance, U. S. Pat. Nos.
8 5,272,236 and 5,278,272; Randall (Rev. Macromol. Chem. Phys. , C29 (2 &3), p. 285-
9 297), which discusses the measurement of long chain branching using ¹³C nuclear
10 magnetic resonance spectroscopy, Zimm, G. H. and Stockmayer, W. H., J. Chem. Phys.,
11 17, 1301 (1949); and Rudin, A., Modern Methods of Polymer Characterization, John
12 Wiley & Sons, New York (1991) pp. 103-112, which discuss the use of gel permeation
13 chromatography coupled with a low angle laser light scattering detector ("GPC-LALLS")
14 and gel permeation chromatography coupled with a differential viscometer detector
15 ("GPC-DV").

16 The homogeneous linear or substantially linear ethylene polymer will be an
17 interpolymer of ethylene with at least one α -olefin. When ethylene propylene diene
18 terpolymers ("EPDM's") are prepared, the dienes are typically non-conjugated dienes
19 having from 6 to 15 carbon atoms. Representative examples of suitable non-conjugated
20 dienes that may be used to prepare the terpolymers include:

- 21 (a) Straight chain acyclic dienes such as 1,4-hexadiene; 1,5-heptadiene; and 1,6-
22 octadiene;
- 23 (b) Branched chain acyclic dienes such as 5-methyl-1,4-hexadiene; 3,7-dimethyl-1,6-
24 octadiene; and 3,7-dimethyl-1,7-octadiene;
- 25 (c) Single ring alicyclic dienes such as 4-vinylcyclohexene; 1-allyl-4-isopropylidene
26 cyclohexane; 3-allylcyclopentene; 4-allylcyclohexene; and 1-isopropenyl-4-
27 butenylcyclohexene;
- 28 (d) Multi-ring alicyclic fused and bridged ring dienes such as dicyclopentadiene;
29 alkenyl, alkylidene, cycloalkenyl, and cycloalkylidene norbornenes, such as 5-
30 methylene-2-norbornene; 5-methylene-6-methyl-2-norbornene; 5-methylene-6,6-

1 dimethyl-2-norbornene; 5-propenyl-2-norbornene; 5-(3-cyclopentenyl)-2-
2 norbornene; 5-ethylidene-2-norbornene; 5-cyclohexylidene-2-norbornene; etc.

3 The preferred dienes are selected from the group consisting of 1,4-hexadiene;
4 dicyclopentadiene; 5-ethylidene-2-norbornene; 5-methylene-2-norbornene; 7-methyl-1,6
5 octadiene; 4-vinylcyclohexene; etc. One preferred conjugated diene, which may be
6 employed is piperylene.

7 Most preferred are interpolymers of ethylene with at least one C₃ -C₃₀ α -olefins
8 (for instance, propylene, isobutylene, 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene,
9 and 1-octene), with interpolymers of ethylene with at least one C₄ - C₂₀ α -olefin,
10 particularly at least one C₇ -C₃₀ α -olefin, being most preferred.

11 The SCBDI or CDBI for the narrow composition distribution homogeneous
12 interpolymers used in the present invention is density (and to a lesser extent molecular
13 weight) dependent. For polymers having densities less than 0.898 g/cm³, the CDBI
14 values, (as defined in US Pat No. 5,548,014), are less than 70%. For interpolymers
15 having densities greater than or equal to 0.898 g/cm³ the CDBI values, (as defined in US
16 Pat No. 5,548,014), are greater than or equal to 70%.

17 The homogeneous interpolymer used in the present invention is a homogeneous
18 polymer of ethylene with at least one ethylenically unsaturated monomer, conjugated or
19 nonconjugated diene, polyene, etc.

20 Homogeneously branched linear ethylene/ α -olefin interpolymers may be prepared
21 using polymerization processes (such as is described by Elston in U.S. Pat. No.
22 3,645,992) which provide a homogeneous short chain branching distribution. In his
23 polymerization process, Elston uses soluble vanadium catalyst systems to make such
24 polymers. However, others such as Mitsui Petrochemical Company and Exxon Chemical
25 Company have used so-called single site metallocene catalyst systems to make polymers
26 having a homogeneous linear structure. Homogeneous linear ethylene/ α -olefin
27 interpolymers are currently available from Mitsui Petrochemical Company under the
28 tradename "TAFMERTM" and from Exxon Chemical Company under the tradename
29 "EXACTTM".

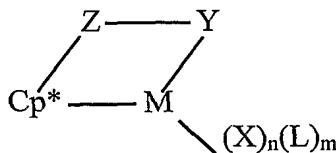
1 Substantially linear ethylene/ α -olefin interpolymers are available from The Dow
2 Chemical Company as AFFINITYTM polyolefin plastomers. Substantially linear
3 ethylene/ α -olefin interpolymers may be prepared in accordance with the techniques
4 described in U.S. Pat. No. 5,272,236 and in U.S. Pat. No. 5,278,272, the entire contents
5 of both of which are herein incorporated by reference.

6 The present invention is a polymer composition, derived from ethylene and alpha
7 olefin, which can be used as an alternative to conventional hot melt adhesives that are
8 subsequently used to bond articles, yet which composition yields adhesive properties
9 similar to adhesives containing polymer, wax and tackifier.

10 The present inventors have discovered that use of a specific type of homogeneous
11 interpolymers can unexpectedly be used by itself or in combination with a tackifier to
12 produce commercially acceptable hot melt adhesives. The present invention is a hot melt
13 adhesive comprising a specific synthetic interpolymers that, when combined with a
14 suitable tackifier, can be used as an alternative to hot melt adhesive formulations that
15 incorporate a three-component wax, polymer and tackifier mixture.

16 The homogenous interpolymers used in the hot melt adhesive formulations of the
17 present invention may be prepared using the constrained geometry catalysts disclosed in
18 U.S. Patents No. 5,064,802, No. 5,132,380, No. 5,703,187, No. 6,034,021, EP 0 468 651,
19 EP 0 514 828, WO 93/19104, and WO 95/00526, all of which are incorporated by
20 references herein in their entirety. Another suitable class of catalysts is the metallocene
21 catalysts disclosed in U.S. Patents No. 5,044,438; No. 5,057,475; No. 5,096,867; and No.
22 5,324,800, all of which are incorporated by reference herein in their entirety. It is noted
23 that constrained geometry catalysts may be considered as metallocene catalysts, and both
24 are sometimes referred to in the art as single-site catalysts.

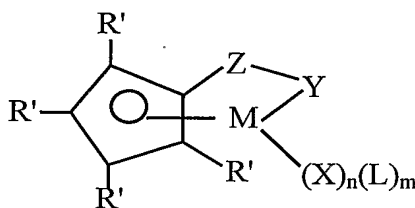
25 For example, catalysts may be selected from the metal coordination complexes
26 corresponding to the formula:



Formula I

wherein: M is a metal of group 3, 4-10, or the lanthanide series of the periodic table of the elements; Cp* is a cyclopentadienyl or substituted cyclopentadienyl group bound in an η^5 bonding mode to M; Z is a moiety comprising boron, or a member of group 14 of the periodic table of the elements, and optionally sulfur or oxygen, the moiety having up to 40 non-hydrogen atoms, and optionally Cp* and Z together form a fused ring system; X independently each occurrence is an anionic ligand group, said X having up to 30 non-hydrogen atoms; n is 2 less than the valence of M when Y is anionic, or 1 less than the valence of M when Y is neutral; L independently each occurrence is a neutral Lewis base ligand group, said L having up to 30 non-hydrogen atoms; m is 0, 1, 2, 3, or 4; and Y is an anionic or neutral ligand group bonded to Z and M comprising nitrogen, phosphorus, oxygen or sulfur and having up to 40 non-hydrogen atoms, optionally Y and Z together form a fused ring system.

Suitable catalysts may also be selected from the metal coordination complex which corresponds to the formula:



Formula II

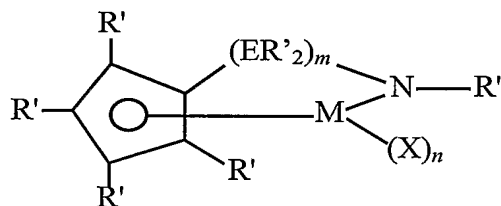
wherein R' each occurrence is independently selected from the group consisting of hydrogen, alkyl, aryl, silyl, germyl, cyano, halo and combinations thereof having up to 20 non-hydrogen atoms; X each occurrence independently is selected from the group consisting of hydride, halo, alkyl, aryl, silyl, germyl, aryloxy, alkoxy, amide, siloxy, and combinations thereof having up to 20 non-hydrogen atoms; L independently each occurrence is a neutral Lewis base ligand having up to 30 non-hydrogen atoms; Y is —O—, —S—, —NR*—, —PR*—, or a neutral two electron donor ligand selected from the group consisting of OR*, SR*, NR*₂, PR*₂; M, n, and m are as previously defined; and Z is SIR*₂, CR*₂, SiR*₂SiR*₂, CR*₂CR*₂, CR*=CR*, CR*₂SiR*₂, GeR*₂, BR*, BR*₂; wherein: R* each occurrence is independently selected from the group consisting of hydrogen, alkyl, aryl, silyl, halogenated alkyl, halogenated aryl groups having up to 20

1 non-hydrogen atoms, and mixtures thereof, or two or more R* groups from Y, Z, or both
 2 Y and Z form a fused ring system.

3 It should be noted that whereas formula I and the following formulas indicate a
 4 monomeric structure for the catalysts, the complex may exist as a dimer or higher
 5 oligomer.

6 Further preferably, at least one of R', Z, or R* is an electron donating moiety.
 7 Thus, highly preferably Y is a nitrogen or phosphorus containing group corresponding to
 8 the formula —N(R''')— or —P(R''')—, wherein R''' is C₁₋₁₀ alkyl or aryl, i.e., an amido
 9 or phosphido group.

10 Additional catalysts may be selected from the amidosilane- or amidoalkanediyl-
 11 compounds corresponding to the formula:



17 Formula III

18 wherein: M is titanium, zirconium or hafnium, bound in an η^5 bonding mode to the
 19 cyclopentadienyl group; R' each occurrence is independently selected from the group
 20 consisting of hydrogen, silyl, alkyl, aryl and combinations thereof having up to 10 carbon
 21 or silicon atoms; E is silicon or carbon; X independently each occurrence is hydride, halo,
 22 alkyl, aryl, aryloxy or alkoxy of up to 10 carbons; m is 1 or 2; and n is 1 or 2 depending
 23 on the valence of M.

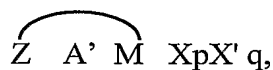
24 Examples of the above metal coordination compounds include, but are not limited
 25 to, compounds in which the R' on the amido group is methyl, ethyl, propyl, butyl, pentyl,
 26 hexyl, (including isomers), norbornyl, benzyl, phenyl, etc.; the cyclopentadienyl group is
 27 cyclopentadienyl, indenyl, tetrahydroindenyl, fluorenyl, octahydrofluorenyl, etc.; R' on
 28 the foregoing cyclopentadienyl groups each occurrence is hydrogen, methyl, ethyl,
 29 propyl, butyl, pentyl, hexyl, (including isomers), norbornyl, benzyl, phenyl, etc.; and X is

chloro, bromo, iodo, methyl, ethyl, propyl, butyl, pentyl, hexyl, (including isomers), norbornyl, benzyl, phenyl, etc.

Specific compounds include, but are not limited to, (tertbutylamido)(tetramethyl- η^5 -cyclopentadienyl)-1,2-ethanediylzirconium dimethyl, (tert-butylamido) (tetramethyl- η^5 -cyclopentadienyl)-1,2-ethanediyltitanium dimethyl, (methylamido) (tetramethyl- η^5 -cyclopentadienyl)-1,2-ethanediylzirconium dichloride, (methylamido)(tetramethyl- η^5 -cyclopentadienyl)-1,2-ethanediyltitanium dichloride, (ethylamido)(tetramethyl- η^5 -cyclopentadienyl)-methylenetitanium dichloro, (tertbutylamido)diphenyl(tetramethyl- η^5 -cyclopentadienyl)-silane zirconium dibenzyl, (benzylamido)dimethyl-(tetramethyl- η^5 -cyclopentadienyl) ilanetitaniumdichloride, phenylphosphido)dimethyl(tetramethyl- η^5 -cyclopentadienyl) silane zirconium dibenzyl, and the like.

Another suitable class of catalysts is substituted indenyl containing metal complexes as disclosed in U.S. Patents No. 5,965,756 and No. 6,015,868, which are incorporated by reference herein in their entirety. Other catalysts are disclosed in copending applications: U.S. Application Serial No. 09/230,185; and No. 09/715,380, and U.S. Provisional Application Serial No. 60/215,456; No. 60/170,175, and No. 60/393,862. The disclosures of all of the preceding patent applications are incorporated by reference herein in their entirety. These catalysts tend to have a higher molecular weight capability.

One class of the above catalysts is the indenyl containing metal wherein:



Formula IV

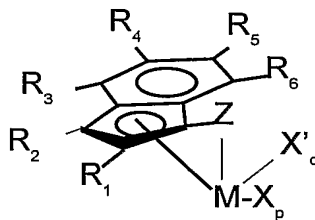
M is titanium, zirconium or hafnium in the +2, +3 or +4 formal oxidation state;

A' is a substituted indenyl group substituted in at least the 2 or 3 position with a group selected from hydrocarbyl, fluoro-substituted hydrocarbyl, hydrocarbyloxy-substituted hydrocarbyl, dialkylamino- substituted hydrocarbyl, silyl, germyl and mixtures thereof, the group containing up to 40 non-hydrogen atoms, and the A' further being covalently bonded to M by means of a divalent Z group; Z is a divalent moiety

bound to both A' and M via σ -bonds, the Z comprising boron, or a member of Group 14 of the Periodic Table of the Elements, and also comprising nitrogen, phosphorus, sulfur or oxygen; X is an anionic or dianionic ligand group having up to 60 atoms exclusive of the class of ligands that are cyclic, delocalized, π -bound ligand groups; X' independently each occurrence is a neutral Lewis base, having up to 20 atoms; p is 0, 1 or 2, and is two less than the formal oxidation state of M, with the proviso that when X is a dianionic ligand group, p is 1; and q is 0, 1 or 2.

The above complexes may exist as isolated crystals optionally in pure form or as a mixture with other complexes, in the form of a solvated adduct, optionally in a solvent, especially an organic liquid, as well as in the form of a dimer or chelated derivative thereof, wherein the chelating agent is an organic material, preferably a neutral Lewis base, especially a trihydrocarbylamine, trihydrocarbylphosphine, or halogenated derivative thereof.

Preferred catalysts are complexes corresponding to the formula:

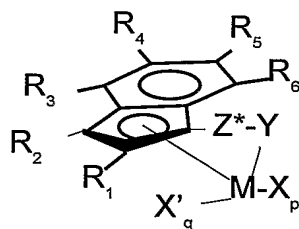


Formula V

wherein R₁ and R₂ independently are groups selected from hydrogen, hydrocarbyl, perfluoro substituted hydrocarbyl, silyl, germyl and mixtures thereof, the group containing up to 20 non-hydrogen atoms, with the proviso that at least one of R₁ or R₂ is not hydrogen; R₃, R₄, R₅, and R₆ independently are groups selected from hydrogen, hydrocarbyl, perfluoro substituted hydrocarbyl, silyl, germyl and mixtures thereof, the group containing up to 20 non-hydrogen atoms; M is titanium, zirconium or hafnium; Z is a divalent moiety comprising boron, or a member of Group 14 of the Periodic Table of the Elements, and also comprising nitrogen, phosphorus, sulfur or oxygen, the moiety having up to 60 non-hydrogen atoms; p is 0, 1 or 2; q is zero or one; with the proviso that: when p is 2, q is zero, M is in the +4 formal oxidation state, and X is an anionic

ligand selected from the group consisting of halide, hydrocarbyl, hydrocarbyloxy, di(hydrocarbyl)amido, di(hydrocarbyl)phosphido, hydrocarbyl sulfido, and silyl groups, as well as halo-, di(hydrocarbyl)amino-, hydrocarbyloxy- and di(hydrocarbyl)phosphino-substituted derivatives thereof, the X group having up to 20 non-hydrogen atoms, when p is 1, q is zero, M is in the +3 formal oxidation state, and X is a stabilizing anionic ligand group selected from the group consisting of allyl, 2-(N,N-dimethylaminomethyl)phenyl, and 2-(N,N-dimethyl)-aminobenzyl, or M is in the +4 formal oxidation state, and X is a divalent derivative of a conjugated diene, M and X together forming a metallocyclopentene group, and when p is 0, q is 1, M is in the +2 formal oxidation state, and X' is a neutral, conjugated or non-conjugated diene, optionally substituted with one or more hydrocarbyl groups, the X' having up to 40 carbon atoms and forming a π -complex with M.

More preferred catalysts are complexes corresponding to the formula:



Formula VI

wherein: R_1 and R_2 are hydrogen or C_{1-6} alkyl, with the proviso that at least one of R_1 or R_2 is not hydrogen; R_3 , R_4 , R_5 , and R_6 independently are hydrogen or C_{1-6} alkyl; M is titanium; Y is $—O—$, $—S—$, $—NR^*—$, $—PR^*—$; Z^* is SiR^{*}_2 , CR^{*}_2 , $SiR^{*}_2SiR^{*}_2$, $CR^{*}_2CR^{*}_2$, $CR^{*}=CR^{*}$, $CR^{*}_2SiR^{*}_2$, or GeR^{*}_2 ; R^* each occurrence is independently hydrogen, or a member selected from hydrocarbyl, hydrocarbyloxy, silyl, halogenated alkyl, halogenated aryl, and combinations thereof, the R^* having up to 20 non-hydrogen atoms, and optionally, two R^* groups from Z (when R^* is not hydrogen), or an R^* group from Z and an R^* group from Y form a ring system; p is 0, 1 or 2; q is zero or one; with the proviso that: when p is 2, q is zero, M is in the +4 formal oxidation state, and X is independently each occurrence methyl or benzyl, when p is 1, q is zero, M is in the +3 formal oxidation state, and X is 2-(N,N-dimethyl)aminobenzyl; or M is in the +4 formal

1 oxidation state and X is 1,4-butadienyl, and when p is 0, q is 1, M is in the +2 formal
2 oxidation state, and X' is 1,4-diphenyl-1,3-butadiene or 1,3-pentadiene. The latter diene
3 is illustrative of unsymmetrical diene groups that result in production of metal complexes
4 that are actually mixtures of the respective geometrical isomers.

5 Other catalysts, cocatalysts, catalyst systems, and activating techniques which
6 may be used in the practice of the invention disclosed herein may include those disclosed
7 in; US Patent 5,616,664, WO 96/23010, published on August 1, 1996, WO 99/14250,
8 published March 25, 1999, WO 98/41529, published September 24, 1998, WO 97/42241,
9 published November 13, 1997, WO 97/42241, published November 13, 1997, those
10 disclosed by Scollard, et al., in J. Am. Chem. Soc 1996, 118, 10008 - 10009, EP 0 468
11 537 B1, published November 13, 1996, WO 97/22635, published June 26, 1997, EP 0
12 949 278 A2, published October 13, 1999; EP 0 949 279 A2, published October 13, 1999;
13 EP 1 063 244 A2, published December 27, 2000; US Patent 5,408,017; US Patent
14 5,767,208; US Patent 5,907,021; WO 88/05792, published August 11, 1988;
15 WO88/05793, published August 11, 1988; WO 93/25590, published December 23,
16 1993; US Patent 5,599,761; US Patent 5,218,071; WO 90/07526, published July 12,
17 1990; US Patent 5,972,822; US Patent 6,074,977; US Patent 6,013,819; US Patent
18 5,296,433; US Patent 4,874,880; US Patent 5,198,401; US Patent 5,621,127; US Patent
19 5,703,257; US Patent 5,728,855; US Patent 5,731,253; US Patent 5,710,224; US Patent
20 5,883,204; US Patent 5,504,049; US Patent 5,962,714; US Patent 5,965,677; US Patent
21 5,427,991; WO 93/21238, published October 28, 1993; WO 94/03506, published
22 February 17, 1994; WO 93/21242, published October 28, 1993; WO 94/00500, published
23 January 6, 1994, WO 96/00244, published January 4, 1996, WO 98/50392, published
24 November 12, 1998; Wang, et al., Organometallics 1998, 17, 3149-3151; Younkin, et al.,
25 Science 2000, 287, 460-462, Chen and Marks, Chem. Rev. 2000, 100, 1391-1434, Alt
26 and Koppl, Chem. Rev. 2000, 100, 1205-1221; Resconi, et al., Chem. Rev. 2000, 100,
27 1253-1345; Ittel, et al., ChemRev. 2000, 100, 1169-1203; Coates, Chem. Rev., 2000, 100,
28 1223-1251; WO 96/13530, published May 9, 1996; all of which patents and publications
29 are herein incorporated by reference in their entirety. Also useful are those catalysts,
30 cocatalysts, and catalyst systems disclosed in USSN 09/230,185, filed January 15, 1999;
31 US Patent 5,965,756; US 6,150,297; USSN 09/715,380, filed November 17, 2000; ; all

1 of which patents and publications are herein incorporated by reference in their entirety.
2 In addition, methods for preparing the aforementioned catalysts are described, for
3 example, in U.S. Patent No. 6,015,868, the entire contents of which are herein
4 incorporated by reference.

5 **Cocatalysts:**

6 The above-described catalysts may be rendered catalytically active by
7 combination with an activating cocatalyst or by use of an activating technique. Suitable
8 activating cocatalysts for use herein include, but are not limited to, polymeric or
9 oligomeric alumoxanes, especially methylalumoxane, triisobutyl aluminum modified
10 methylalumoxane, or isobutylalumoxane; neutral Lewis acids, such as C₁₋₃₀ hydrocarbyl
11 substituted Group 13 compounds, especially tri(hydrocarbyl)aluminum- or
12 tri(hydrocarbyl)boron compounds and halogenated (including perhalogenated)
13 derivatives thereof, having from 1 to 30 carbons in each hydrocarbyl or halogenated
14 hydrocarbyl group, more especially perfluorinated tri(aryl)boron and perfluorinated
15 tri(aryl)aluminum compounds, mixtures of fluoro-substituted(aryl)boron compounds with
16 alkyl-containing aluminum compounds, especially mixtures of
17 tris(pentafluorophenyl)borane with trialkylaluminum or mixtures of
18 tris(pentafluorophenyl)borane with alkylalumoxanes, more especially mixtures of
19 tris(pentafluorophenyl)borane with methylalumoxane and mixtures of
20 tris(pentafluorophenyl)borane with methylalumoxane modified with a percentage of
21 higher alkyl groups (MMAO), and most especially tris(pentafluorophenyl)borane and
22 tris(pentafluorophenyl)aluminum; non-polymeric, compatible, non-coordinating, ion
23 forming compounds (including the use of such compounds under oxidizing conditions),
24 especially the use of ammonium-, phosphonium-, oxonium-, carbonium-, silylium- or
25 sulfonium- salts of compatible, non-coordinating anions, or ferrocenium salts of
26 compatible, non-coordinating anions; bulk electrolysis and combinations of the foregoing
27 activating cocatalysts and techniques. The foregoing activating cocatalysts and activating
28 techniques have been previously taught with respect to different metal complexes in the
29 following references: EP-A-277,003, US-A-5,153,157, US-A-5,064,802, EP-A-468,651
30 (equivalent to U. S. Serial No. 07/547,718), EP-A-520,732 (equivalent to U. S. Serial No.
31 07/876,268), and EP-A-520,732 (equivalent to U. S. Serial Nos. 07/884,966 filed May 1,

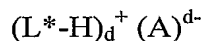
1992). The disclosures of the all of the preceding patents or patent applications are incorporated by reference herein in their entirety.

Combinations of neutral Lewis acids, especially the combination of a trialkyl aluminum compound having from 1 to 4 carbons in each alkyl group and a halogenated tri(hydrocarbyl)boron compound having from 1 to 20 carbons in each hydrocarbyl group, especially tris(pentafluorophenyl)borane, further combinations of such neutral Lewis acid mixtures with a polymeric or oligomeric alumoxane, and combinations of a single neutral Lewis acid, especially tris(pentafluorophenyl)borane with a polymeric or oligomeric alumoxane are especially desirable activating cocatalysts. It has been observed that the most efficient catalyst activation using such a combination of tris(pentafluoro-phenyl)borane/alumoxane mixture occurs at reduced levels of alumoxane. Preferred molar ratios of Group 4 metal complex:tris(pentafluoro-phenyl)borane:alumoxane are from 1:1:1 to 1:5:10, more preferably from 1:1:1 to 1:3:5. Such efficient use of lower levels of alumoxane allows for the production of olefin polymers with high catalytic efficiencies using less of the expensive alumoxane cocatalyst. Additionally, polymers with lower levels of aluminum residue, and hence greater clarity, are obtained.

Suitable ion forming compounds useful as cocatalysts in some embodiments of the invention comprise a cation which is a Bronsted acid capable of donating a proton, and a compatible, non-coordinating anion, A⁻. As used herein, the term "non-coordinating" means an anion or substance which either does not coordinate to the Group 4 metal containing precursor complex and the catalytic derivative derived therefrom, or which is only weakly coordinated to such complexes thereby remaining sufficiently labile to be displaced by a neutral Lewis base. A non-coordinating anion specifically refers to an anion which, when functioning as a charge balancing anion in a cationic metal complex, does not transfer an anionic substituent or fragment thereof to the cation thereby forming neutral complexes during the time which would substantially interfere with the intended use of the cationic metal complex as a catalyst.. "Compatible anions" are anions which are not degraded to neutrality when the initially formed complex decomposes and are non-interfering with desired subsequent polymerization or other uses of the complex.

Preferred anions are those containing a single coordination complex comprising a charge-bearing metal or metalloid core which anion is capable of balancing the charge of the active catalyst species (the metal cation) which may be formed when the two components are combined. Also, the anion should be sufficiently labile to be displaced by olefinic, diolefinic and acetylenically unsaturated compounds or other neutral Lewis bases such as ethers or nitriles. Suitable metals include, but are not limited to, aluminum, gold and platinum. Suitable metalloids include, but are not limited to, boron, phosphorus, and silicon. Compounds containing anions which comprise coordination complexes containing a single metal or metalloid atom are, of course, known in the art and many, particularly such compounds containing a single boron atom in the anion portion, are available commercially.

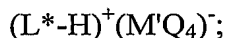
Preferably such cocatalysts may be represented by the following general formula:



Formula VII

wherein L^* is a neutral Lewis base; $(L^*-H)^+$ is a Bronsted acid; A^{d-} is an anion having a charge of $d-$, and d is an integer from 1 to 3. More preferably A^{d-} corresponds to the formula: $[M'Q_4]^-$, wherein M' is boron or aluminum in the +3 formal oxidation state; and Q independently each occurrence is selected from hydride, dialkylamido, halide, hydrocarbyl, hydrocarbyloxy, halosubstituted-hydrocarbyl, halosubstituted hydrocarbyloxy, and halo-substituted silylhydrocarbyl radicals (including perhalogenated hydrocarbyl- perhalogenated hydrocarbyloxy- and perhalogenated silylhydrocarbyl radicals), the Q having up to 20 carbons with the proviso that in not more than one occurrence is Q halide. Examples of suitable hydrocarbyloxy Q groups are disclosed in U. S. Patent 5,296,433.

In a more preferred embodiment, d is one, that is, the counter ion has a single negative charge and is A^- . Activating cocatalysts comprising boron which are particularly useful in the preparation of catalysts of this invention may be represented by the following general formula:



Formula VIII

wherein L* is as previously defined; M' is boron or aluminum in a formal oxidation state of 3; and Q is a hydrocarbyl-, hydrocarbyloxy-, fluorinated hydrocarbyl-, fluorinated hydrocarbyloxy-, or fluorinated silylhydrocarbyl- group of up to 20 non-hydrogen atoms, with the proviso that in not more than one occasion is Q hydrocarbyl. Most preferably, Q in each occurrence is a fluorinated aryl group, especially a pentafluorophenyl group. Preferred (L*-H)⁺ cations are N,N-dimethylanilinium, N,N-di(octadecyl)anilinium, di(octadecyl)methylammonium, methylbis(hydrogenated tallowyl)ammonium, and tributylammonium.

Illustrative, but not limiting, examples of boron compounds which may be used as an activating cocatalyst are tri-substituted ammonium salts such as: trimethylammonium tetrakis(pentafluorophenyl) borate; triethylammonium tetrakis(pentafluorophenyl) borate; tripropylammonium tetrakis (pentafluorophenyl) borate; tri(n-butyl)ammonium tetrakis(pentafluorophenyl) borate; tri(sec-butyl)ammonium tetrakis(pentafluorophenyl) borate; N,N-dimethylanilinium tetrakis (pentafluorophenyl) borate; N,N-dimethylanilinium n-butyltris(pentafluorophenyl) borate; N,N-dimethylanilinium benzyltris(pentafluorophenyl) borate; N,N-dimethylanilinium tetrakis(4-(t-butyl)dimethylsilyl)-2, 3, 5, 6-tetrafluorophenyl) borate; N,N-dimethylanilinium tetrakis(4-(triisopropylsilyl)-2, 3, 5, 6-tetrafluorophenyl) borate; N,N-dimethylanilinium pentafluoro phenoxytris(pentafluorophenyl) borate; N,N-diethylanilinium tetrakis(pentafluorophenyl) borate; N,N-dimethyl-2,4,6-trimethylanilinium tetrakis(pentafluorophenyl) borate; trimethylammonium tetrakis(2,3,4,6-tetrafluorophenyl)borate; triethylammonium tetrakis(2,3,4,6-tetrafluorophenyl) borate; tripropylammonium tetrakis(2,3,4,6-tetrafluorophenyl) borate; tri(n-butyl)ammonium tetrakis(2,3,4,6-tetrafluorophenyl) borate, dimethyl(t-butyl)ammonium tetrakis(2,3,4,6-tetra fluorophenyl) borate; N,N-dimethylanilinium tetrakis(2,3,4,6-tetrafluorophenyl) borate; N,N-diethylanilinium tetrakis (2,3,4,6-tetrafluorophenyl) borate; and N,N-dimethyl-2,4,6-trimethylanilinium tetrakis(2,3,4,6-tetrafluorophenyl) borate; dialkyl ammonium salts such as: di-(i-propyl)ammonium tetrakis(pentafluorophenyl) borate, and dicyclohexylammonium tetrakis(pentafluorophenyl) borate; tri-substituted phosphonium

1 salts such as: triphenylphosphonium tetrakis (pentafluorophenyl) borate, tri(o-
2 tolyl)phosphonium tetrakis(pentafluorophenyl) borate, and tri(2,6-
3 dimethylphenyl)phosphonium tetrakis(pentafluorophenyl) borate; di-substituted oxonium
4 salts such as: diphenyloxonium tetrakis(pentafluorophenyl) borate, di(o-tolyl)oxonium
5 tetrakis (pentafluorophenyl) borate, and di(2,6-dimethylphenyl)oxonium
6 tetrakis(pentafluorophenyl) borate; di-substituted sulfonium salts such as:
7 diphenylsulfonium tetrakis(pentafluorophenyl) borate, di(o-tolyl)sulfonium
8 tetrakis(pentafluorophenyl) borate, and bis(2,6-dimethylphenyl) sulfonium
9 tetrakis(pentafluorophenyl) borate.

10 Preferred silylium salt activating cocatalysts include, but are not limited to,
11 trimethylsilylium tetrakis(pentafluorophenyl)borate, triethylsilylium tetrakis(pentafluoro-
12 phenyl)borate and ether substituted adducts thereof. Silylium salts have been previously
13 generically disclosed in J. Chem. Soc. Chem. Comm., 1993, 383-384, as well as Lambert,
14 J. B., et al., Organometallics, 1994, 13, 2430-2443. The use of the above silylium salts as
15 activating cocatalysts for addition polymerization catalysts is disclosed in U.S. Patent No.
16 5,625,087, which is incorporated by reference herein in its entirety. Certain complexes of
17 alcohols, mercaptans, silanols, and oximes with tris(pentafluorophenyl)borane are also
18 effective catalyst activators and may be used in embodiments of the invention. Such
19 cocatalysts are disclosed in U.S. Patent No. 5,296,433, which is also incorporated by
20 reference herein in its entirety.

21 The catalyst system may be prepared as a homogeneous catalyst by addition of
22 the requisite components to a solvent in which polymerization will be carried out by
23 solution polymerization procedures. The catalyst system may also be prepared and
24 employed as a heterogeneous catalyst by adsorbing the requisite components on a catalyst
25 support material such as silica gel, alumina or other suitable inorganic support material.
26 When prepared in heterogeneous or supported form, it is preferred to use silica as the
27 support material.

28 At all times, the individual ingredients, as well as the catalyst components, should
29 be protected from oxygen and moisture. Therefore, the catalyst components and catalysts
30 should be prepared and recovered in an oxygen and moisture free atmosphere.

1 Preferably, therefore, the reactions are performed in the presence of a dry, inert gas such
2 as, for example, nitrogen or argon.

3 The molar ratio of metal complex: activating cocatalyst employed preferably
4 ranges from 1 : 1000 to 2 : 1, more preferably from 1 : 5 to 1.5 : 1, most preferably from
5 1 : 2 to 1 : 1. In the preferred case in which a metal complex is activated by
6 trispentafluorophenylborane and triisobutylaluminum modified methylalumoxane, the
7 titanium:boron:aluminum molar ratio is typically from 1 : 10 : 50 to 1 : 0.5 : 0.1, most
8 typically from about 1 : 3 : 5.

9 In general, the polymerization may be accomplished at conditions for Ziegler-
10 Natta or Kaminsky-Sinn type polymerization reactions, that is, reactor pressures ranging
11 from atmospheric to 3500 atmospheres (34.5 kPa). The reactor temperature should be
12 greater than 80°C, typically from 100°C to 250°C, and preferably from 100°C to 150°C,
13 with higher reactor temperatures, that is, reactor temperatures greater than 100°C
14 generally favoring the formation of lower molecular weight polymers.

15 Generally the polymerization process is carried out with a differential pressure of
16 ethylene of from 10 to 1000 psi (70 to 7000 kPa), most preferably from 40 to 60 psi (300
17 to 400 kPa). The polymerization is generally conducted at a temperature of from 80 to
18 250°C, preferably from 90 to 170 °C, and most preferably from greater than 95 to 160 °C.

19 In most polymerization reactions the molar ratio of catalyst:polymerizable
20 compounds employed is from 10^{-12} :1 to 10^{-1} :1, more preferably from 10^{-9} :1 to 10^{-5} :1.

21 Solution polymerization conditions utilize a solvent for the respective components
22 of the reaction. Preferred solvents include mineral oils and the various hydrocarbons
23 which are liquid at reaction temperatures. Illustrative examples of useful solvents include
24 alkanes such as pentane, isopentane, hexane, heptane, octane and nonane, as well as
25 mixtures of alkanes including kerosene and Isopar E™, available from Exxon Chemicals
26 Inc.; cycloalkanes such as cyclopentane and cyclohexane; and aromatics such as benzene,
27 toluene, xylenes, ethylbenzene and diethylbenzene.

28 The solvent will be present in an amount sufficient to prevent phase separation in
29 the reactor. As the solvent functions to absorb heat, less solvent leads to a less adiabatic

1 reactor. The solvent:ethylene ratio (weight basis) will typically be from 2.5 : 1 to 12 :
2 1, beyond which point catalyst efficiency suffers. The most typical solvent:ethylene ratio
3 (weight basis) is in the range of from 5 : 1 to 10 : 1.

4 The polymerization may be carried out as a batchwise or a continuous
5 polymerization process, with continuous polymerizations processes being required for the
6 preparation of substantially linear polymers. In a continuous process, ethylene,
7 comonomer, and optionally solvent and diene are continuously supplied to the reaction
8 zone and polymer product continuously removed therefrom.

9 b) Tackifier Component

10 Addition of tackifier is desirable to allow for bonding prior to solidifying or
11 setting of the adhesive. An example of this is in high-speed cereal box sealing operations
12 where the overlapping flaps of the box need to adhere to one another while the hot melt
13 adhesive solidifies.

14 Tackifying resins useful in the present invention include aliphatic, cycloaliphatic
15 and aromatic hydrocarbons and modified hydrocarbons and hydrogenated versions;
16 terpenes and modified terpenes and hydrogenated versions; and rosins and rosin
17 derivatives and hydrogenated versions; and mixtures thereof. These tackifying resins
18 have a ring and ball softening point from 70°C. to 150°C, and will typically have a
19 viscosity at 350°F (177°C), as measured using a Brookfield viscometer, of no more than
20 2000 centipoise. They are also available with differing levels of hydrogenation, or
21 saturation, which is another commonly used term. Useful examples include EastotacTM
22 H-100, H-115 and H-130 from Eastman Chemical Co. in Kingsport, Tenn., which are
23 partially hydrogenated cycloaliphatic petroleum hydrocarbon resins with softening points
24 of 100°C, 115°C and 130°C., respectively. These are available in the E grade, the R
25 grade, the L grade and the W grade, indicating differing levels of hydrogenation with E
26 being the least hydrogenated and W being the most hydrogenated. The E grade has a
27 bromine number of 15, the R grade a bromine number of 5, the L grade a bromine
28 number of 3 and the W grade has a bromine number of 1. EastotacTMH-142R from
29 Eastman Chemical Co. has a softening point of about 140°C. Other useful tackifying
30 resins include EscorezTM5300, 5400 and 5637, partially hydrogenated cycloaliphatic

1 petroleum hydrocarbon resins, and EscorezTM 5600, a partially hydrogenated aromatic
2 modified petroleum hydrocarbon resin all available from Exxon Chemical Co. in
3 Houston, Tex.; WingtackTM Extra, which is an aliphatic, aromatic petroleum hydrocarbon
4 resin available from Goodyear Chemical Co. in Akron, Ohio; HerculiteTM 2100, a
5 partially hydrogenated cycloaliphatic petroleum hydrocarbon resin available from
6 Hercules, Inc. in Wilmington, Del.

7 There are numerous types of rosins and modified rosins available with differing
8 levels of hydrogenation including gum rosins, wood rosins, tall-oil rosins, distilled rosins,
9 dimerized rosins and polymerized rosins. Some specific modified rosins include glycerol
10 and pentaerythritol esters of wood rosins and tall-oil rosins. Commercially available
11 grades include, but are not limited to, SylvatacTM 1103, a pentaerythritol rosin ester
12 available from Arizona Chemical Co., UnitacTM R-100 Lite, a pentaerythritol rosin ester
13 from Union Camp in Wayne, N.J., PermalynTM 305, an erythritol modified wood rosin
14 available from Hercules and Foral 105 which is a highly hydrogenated pentaerythritol
15 rosin ester also available from Hercules. SylvatacTM R-85 and 295 are 85°C and 95°C
16 melt point rosin acids available from Arizona Chemical Co. and Foral AX is a 70°C melt
17 point hydrogenated rosin acid available from Hercules, Inc. Nirez V-2040 is a phenolic
18 modified terpene resin available from Arizona Chemical Co.

19 Another exemplary tackifier, Piccotac 115, has a viscosity at 350°F (177°C) of
20 about 1600 centipoise. Other typical tackifiers have viscosities at 350°F (177°C) of
21 much less than 1600 centipoise, for instance, from 50 to 300 centipoise.

22 Exemplary aliphatic resins include those available under the trade designations
23 EscorezTM, PiccotacTM, MercuresTM, WingtackTM, Hi-RezTM, QuintoneTM, TackirolTM,
24 etc. Exemplary polyterpene resins include those available under the trade designations
25 NirezTM, PiccolyteTM, WingtackTM, ZonarezTM, etc. Exemplary hydrogenated resins
26 include those available under the trade designations EscorezTM, ArkonTM, ClearonTM, etc.
27 Exemplary mixed aliphatic-aromatic resins include those available under the trade
28 designations EscorezTM, RegaliteTM, HercuresTM, ARTM, ImprezTM, NorsoleneTM M,
29 MarukarezTM, ArkonTM M, QuintoneTM, etc. These tackifiers may be employed with the
30 polymers of the present invention, providing they are used at compatible levels. Other

1 tackifiers may be employed, provided they are compatible with the homogeneous linear
2 or substantially linear ethylene/alpha.-olefin interpolpolymer.

3 In certain applications of the present invention it is anticipated the hot melt
4 adhesive will be prepared without the use of a tackifier or with a minimal quantity of
5 tackifier. As tackifiers are malodorous, tend to cause corrosion of mechanical equipment,
6 and cannot be easily separated from recycled paper pulp, hot melt adhesives which
7 minimize the use of tackifiers are advantageous. Moreover, as tackifiers generally
8 undergo degradation at elevated temperatures, hot melt adhesives which minimize the use
9 of tackifiers will exhibit improved thermal stability.

10 Tackifiers added to hot-melt adhesives can be characterized by parameters such as
11 their softening points, specific gravities, or by acid number. A tackifier can be selected
12 from among the variety of tackifiers, as described above but not limited thereto, and from
13 tackifiers characterized by a range of acid numbers, such as acid numbers between 0 and
14 100, more preferably between 0 and 25.8, and most preferably a tackifier having an acid
15 number between 3-10.

16
17 c) Other Additives

18 Adhesives, including those of the present invention may also contain a number of
19 additional components, such as a stabilizer, plasticizer, filler or antioxidant. Among the
20 applicable stabilizers or antioxidants which can be included in the adhesive composition
21 of the present invention are high molecular weight hindered phenols and multifunctional
22 phenols, such as sulfur-containing and phosphorous-containing phenols. Hindered
23 phenols, known to those skilled in the art, may be described as phenolic compounds,
24 which also contain sterically bulky radicals in close proximity to the phenolic hydroxyl
25 group. Specifically, tertiary butyl groups generally are substituted onto the benzene ring
26 in at least one of the ortho positions relative to the phenolic hydroxyl group. The
27 presence of these sterically bulky substituted radicals in the vicinity of the hydroxyl
28 group serves to retard its stretching frequency, and correspondingly, its reactivity. It is
29 this hindrance that provides the stabilizing properties of these phenolic compounds.

30 Representative hindered phenols include; but are not limited to: 2,4,6-trialkylated
31 monohydroxy phenols; 1,3,5-trimethyl-2,4,6-tris-(3,5-di-tert-butyl-4-hydroxybenzyl)-

1 benzene; pentaerythritol tetrakis-3(3,5-di-tert-butyl-4-hydroxyphenyl)-propionate,
2 commercially available under the trademark IRGANOX® 1010; n-octadecyl-3(3,5-di-
3 tert-butyl-4-hydroxyphenyl)-propionate; 4,4'-methylenebis (4-methyl-6-tert-butyl-
4 phenol); 4,4'-thiobis (6-tert-butyl-o-cresol); 2,6-di-tertbutylphenol; 6-(4-
5 hydroxyphenoxy)-2,4-bis(n-octyl-thio)-1,3,5 triazine; 2-(n-octylthio)ethyl 3,5-di-tert-
6 butyl-4-hydroxy-benzoate; di-n-octadecyl 3,5-di-tert-butyl-4-hydroxy-
7 benzylphosphonate; and sorbitol hexa(3,3,5-di-tert-butyl-4-hydroxy-phenyl)-propionate.

8 Antioxidants include, but are not limited to, butylated hydroxy anisole ("BHA")
9 or butylated hydroxy toluene ("BHT") which may also be utilized to render the
10 formulation more thermally stable. These stabilizers and antioxidants are added in
11 amounts ranging approximately 0.01 % to approximately 5% by weight of the
12 formulation.

13 Utilizing known synergists in conjunction with the antioxidants may further
14 enhance the performance of these antioxidants. Some of these known synergists are, for
15 example, thiodipropionate esters and phosphates. Chelating agents and metal
16 deactivators, may also be used. Examples of these compounds include
17 ethylenediaminetetraacetic acid ("EDTA"), and more preferably, its salts, and
18 disalicylalpropylenediamine. Distearylthiodipropionate is particularly useful. When
19 added to the adhesive composition, these stabilizers, if used, are generally present in
20 amounts of about 0.1 to about 1.5 weight percent, and more preferably in the range of
21 about 0.25 to about 1.0 weight percent.

22 The present invention also contemplates the addition of a polymeric additive to
23 the adhesive. The polymeric additive can be selected from the group consisting of
24 ethylene methyl acrylate polymers containing 10 to 28 weight percent by weight methyl
25 acrylate; ethylene acrylic acid copolymers having an acid number of 25 to 150;
26 polyethylene; polypropylene; poly(butene-1-co-ethylene) polymers and low molecular
27 weight and/or low melt index ethylene n-butyl acrylate copolymers. When such a
28 polymeric additive is added, it is present in amounts up to about 15 weight percent by
29 weight of composition.

30 Depending on the contemplated end uses of the adhesive composition, other
31 additives such as plasticizers, pigments and dyestuffs that are conventionally added to

1 hot-melt adhesives may be included. In addition, small amounts of additional
2 (secondary) tackifiers and/or waxes such as microcrystalline waxes, hydrogenated castor
3 oil, styrene-ethylene butyl styrene (SEBS) resins and vinyl acetate modified synthetic
4 waxes may also be incorporated in minor amounts, i.e., up to about 10 weight percent by
5 weight, into the formulations of the present invention. A plasticizer may be used in lieu
6 of, or in combination with, the secondary tackifier to modify viscosity and improve the
7 tack properties of the adhesive composition.

8 A dispersant can also be added to these compositions. The dispersant can be a
9 chemical, which may, by itself, cause the composition to be dispersed from the surface to
10 which it has been applied, for example, under aqueous conditions. The dispersant may
11 also be an agent which when chemically modified, causes the composition to be
12 dispersed from the surface to which it has been applied. As known to those skilled in the
13 art, examples of these dispersants include surfactants, emulsifying agents, and various
14 cationic, anionic or nonionic dispersants. Compounds such as amines, amides and their
15 derivatives are examples of cationic dispersants. Soaps, acids, esters and alcohols are
16 among the known anionic dispersants. The addition of a dispersant may affect the
17 recyclability of products to which a hot-melt adhesive may have been applied.

18 The surfactants can be chosen from a variety of known surface-active agents.
19 These can include nonionic compounds such as ethoxylates available from commercial
20 suppliers. Examples include alcohol ethoxylates, alkylamine ethoxylates, alkylphenol
21 ethoxylates, octylphenol ethoxylates and the like. Other surfactants, such as a number
22 of fatty acid esters may be employed; for example, but not limited to, glycerol esters,
23 polyethyleneglycol esters and sorbitan esters.

24
25 Although the present invention has been described with a certain degree of
26 particularity, it is to be understood that the examples below are merely for purposes of
27 illustrating the present invention, the scope of the present invention is not intended to be
28 defined by the claims.

29
30 Composition and Properties of the Hot Melt Adhesive of the Present Invention

1 The hot melt adhesive of the present invention consists essentially of from about
2 40 to about 100 weight percent, preferably from about 60 to about 85 weight percent,
3 more preferably from about 65 to about 80 weight percent (based on the final weight of
4 the hot melt adhesive) of an ethylene alpha olefin interpolymers, and of from 0 to about 60
5 weight percent, preferably from about 15 to about 40 weight percent, more preferably
6 from about 20 to about 35 weight percent (based on the final weight of the hot melt
7 adhesive) of one or more tackifiers.

8 The ethylene alpha olefin interpolymers component of the hot melt adhesives of
9 the present invention has a density of from about 0.880 to about 0.930 g/cm³, preferably
10 from about 0.890 to about 0.920 g/cm³, more preferably from about 0.8945 to about
11 0.915 g/cm³, even more preferably greater than 0.885 g/cm³, and most preferably greater
12 than 0.895 g/cm³.

13 The ethylene alpha olefin interpolymers component of the hot melt adhesives of
14 the present invention has a number average molecular weight (Mn as measured by GPC)
15 of from about 1,000 to about 9,000, preferably from about 1,250 to about 7,000, more
16 preferably from about 1,500 to about 6000.

17 The ethylene alpha olefin interpolymers component of the hot melt adhesives of
18 the present invention has a Brookfield Viscosity (measured at 300°F) of from about 500
19 to about 7,000 cP, preferably from about 1,000 to about 6,000 cP, more preferably from
20 about 1,500 to about 5000 cP.

21 The hot melt adhesives of the present invention have a Brookfield Viscosity
22 (measured at 350°F) of from about 400 to about 2,000 cP, preferably from about 500 to
23 about 1,400 cP, more preferably from about 750 to about 1,200 cP.

24 The hot melt adhesives of the present invention have a Peel Adhesion Failure
25 Temperature (PAFT) of greater than or equal to 110°F, preferably greater than or equal to
26 115°F, more preferably greater than or equal to 120°F.

27 The hot melt adhesives of the present invention have a Shear Adhesion Failure
28 Temperature (SAFT) of greater than or equal to 140°F, greater than or equal to 150°F,
29 more preferably greater than or equal to 170°F.

1 The hot melt adhesives of the present invention exhibit 100% fiber tear between
2 77°F - 140°F, preferably 100% fiber tear at 35°C - 140°F, most preferably 100% fiber tear
3 at 140°F.

5 PREPARATION OF EXAMPLES

6 Unless otherwise stated, the following examples reference to viscosity was
7 determined in accordance with the following procedure using a Brookfield
8 Laboratories DVII+ Viscometer in disposable aluminum sample chambers. The
9 spindle used is a SC-31 hot-melt spindle, suitable for measuring viscosities in the
10 range of from 10 to 100,000 centipoise. A cutting blade is employed to cut
11 samples into pieces small enough to fit into the 1 inch wide, 5 inches long sample
12 chamber. The sample is placed in the chamber, which is in turn inserted into a
13 Brookfield Thermosel and locked into place with bent needle-nose pliers. The
14 sample chamber has a notch on the bottom that fits the bottom of the Brookfield
15 Thermosel to ensure that the chamber is not allowed to turn when the spindle is
16 inserted and spinning. The sample is heated to the desired temperature, such as
17 300°F or 350°F, with additional sample being added until the melted sample is
18 about 1 inch below the top of the sample chamber. The viscometer apparatus is
19 lowered and the spindle submerged into the sample chamber. Lowering is
20 continued until brackets on the viscometer align on the Thermosel. The
21 viscometer is turned on, and set to a shear rate which leads to a torque reading in
22 the range of 30 to 60 percent. Readings are taken every minute for about 15
23 minutes, or until the values stabilize, which final reading is recorded.

24 Unless otherwise stated, the Shear Adhesion Failure Temperature ("SAFT") test,
25 (a test commonly used to evaluate adhesive performance, and well known to those versed
26 in the industry) were conducted using a standard SAFT test method (ASTM D-4498).
27 SAFT tests were run using a ChemInstruments HT-8 Oven Shear Tester using a 500 gm
28 weight. The tests were started at room temperature (25°C / 77°F) and the temperature
29 increased at the rate of 0.5 degrees C/min. The results were converted and reported in
30 degrees F. The SAFT test measures the temperature at which an adhesive fails.

1
2 Unless otherwise stated, Peel Adhesion Failure Temperature ("PAFT") was
3 conducted according to ASTM D- D4498 (modified for peel mode) using 100 gram
4 weights. PAFT gives a measure of the adherence, when peeled at 180° angle, to a
5 standard steel panel or to other surface of interest for a single-coated tape.

6 Unless otherwise stated % fiber tear on corrugated paper was conducted
7 according to standard industry test methods where a drop of adhesive heated to 350 F is
8 applied on the paper. After 1.5 seconds another paper of a given size (11" x 3") is placed
9 on the adhesive drop and laminated to the base paper. The two sheets are manually pulled
10 apart rapidly and the % fiber tear (FT) estimated.

11 Unless otherwise stated, melting points of the adhesive formulations of the
12 present invention used Differential Scanning Calorimetry ("DSC"). A few milligram of
13 sample are placed into the instrument and the temperature was increased from room
14 temperature to 180°C at 10°C per minute. The sample was then held isothermally at
15 180°C for 3 minutes, and then the temperature was ramped down at 10°C per minute to
16 minus 40°C. The sample was held isothermally at -40°C for 3 minutes. The temperature
17 was then ramped up at 10°C per minute to 150°C.. Crystallinity and melting point data
18 were reported from the second heat curve.

19 Density of the samples is determined in accordance with ASTM D 792.

20 The drop point of the samples is determined in accordance with ASTM 3954
21 (Mettler Drop Point).

22 Unless otherwise stated the evaluation of the adhesive properties of the inventive
23 formulations was conducted by coating onto 45# basis weight kraft paper typically used
24 in the manufacture of cardboard boxes and purchased from National Papers, Minneapolis,
25 Minn.

Table 1 - Commercially Available Materials Used in Evaluations

Ingredient	Supplier
Escorez 5400	ExxonMobil Chemical Company Houston, TX – Cyclical hydrogenated hydrocarbon tackifier resin with softening point of 103° C.
Escorez 5637	ExxonMobil Chemical Company Houston, TX – aromatic modified cycloaliphatic hydrocarbon tackifier resin with softening point of 127-133°C.
Eastotack H 1300W	Eastman Chemical Company Kingsport, TN. ring and ball softening point of 130°C and a Gardner color (molten state) of <1, <i>Eastotac</i> hydrocarbon resins are hydrogenated C ₅ aliphatic hydrocarbon tackifying resins.
Advantra® HL-9250	H.B. Fuller Company St. Paul, MN – formulated adhesive for carton and uncoated corrugated stocks with a viscosity at 325°F of 1,255cps and specific gravity of 0.929 g/cm ³ .
Advantra® HL-9255	H.B. Fuller Company St. Paul, MN – formulated adhesive for wrapper and coated carton stocks with a viscosity at 325°F of 1,140cps and specific gravity of 0.943 g/cm ³ .
BAM Futura 1	IDC – A Division of Ambersil, England – hot melt adhesive for books, magazines, catalogues and directories.
HL-7268	H.B. Fuller Company St. Paul, MN.
HL-2835	H.B. Fuller Company St. Paul, MN – formulated adhesive with moderate speed of set, good flexibility, for bonding a variety of substrates, with a viscosity at 300°F of 2,200cP.
Henkel 80-8488	Henkel Consumer Adhesives Inc. Avon, OH - formulated adhesive for bonding a variety of substrates, with a viscosity at 350°F of 1,080cP.
Henkel 80-8368	Henkel Consumer Adhesives Inc. Avon, OH - formulated adhesive for bonding a variety of substrates, with a viscosity at 350°F of 970cP.
ULTRATHENE® 612-04	EVA resin with 18% VA content, made by Equistar Chemical, LP.
FORAL® 85	Rosin Ester tackifier, made by Hercules.
Wax, 1251/7	Microcrystalline Control supplied by Frank B. Ross Co.
EVA-1	A formulation of 33 wt% ULTRATHENE® 612-04 (18% vinyl acetate co-monomer); 33% FORAL® 85 33% Wax, 1251/7.

Example 1. Ethylene/alpha-Olefin Polymer Preparation Using Single Metallocene Catalyst System

A series of ethylene/ α -olefin interpolymers were also prepared in a 1 gallon, oil jacketed, Autoclave continuously stirred tank reactor (“CSTR”). A magnetically coupled agitator with Lightning A-320 impellers provided the mixing. The reactor ran liquid full

1 at 475 psig (3,275 kPa). Process flow was in at the bottom and out of the top. A heat
2 transfer oil was circulated through the jacket of the reactor to remove some of the heat of
3 reaction. At the exit of the reactor was a Micro-Motion™ flow meter that measured flow
4 and solution density. All lines on the exit of the reactor were traced with 50 psi (344.7
5 kPa) steam and insulated.

6 ISOPAR-E solvent and comonomer were supplied to the reactor at 30 psig
7 pressure. The solvent feed to the reactors was measured by a Micro-Motion™ mass flow
8 meter. A variable speed diaphragm pump controlled the solvent flow rate and increased
9 the solvent pressure to reactor pressure. The comonomer was metered by a Micro-
10 Motion™ mass flow meter and flow controlled by a Research control valve. The
11 comonomer stream was mixed with the solvent stream at the suction of the solvent pump
12 and was pumped to the reactor with the solvent. The remaining solvent was combined
13 with ethylene and (optionally) hydrogen and delivered to the reactor. The ethylene stream
14 was measured by a Micro-Motion™ mass flow meter just prior to the Research valve
15 controlling flow. Three Brooks flow meter/controllers (1 - 200 sccm and 2 - 100sccm)
16 were used to deliver hydrogen into the ethylene stream at the outlet of the ethylene
17 control valve.

18 The ethylene or ethylene / hydrogen mixture combined with the solvent /
19 comonomer stream at ambient temperature. The temperature of the solvent/monomer as it
20 enters the reactor was controlled with two heat exchangers. This stream enters the
21 bottom of the 1 gallon CSTR. The three component catalyst system and its solvent flush
22 also enter the reactor at the bottom but through a different port than the monomer stream.

23 Polymerization was stopped with the addition of catalyst kill into the reactor
24 product line after the meter measuring the solution density. Other polymer additives
25 could be added with the catalyst kill. The reactor effluent stream then entered a post
26 reactor heater that provides additional energy for the solvent removal flash. This flash
27 occurs as the effluent exits the post reactor heater and the pressure is dropped from 475
28 psig down to 10 at the reactor pressure control valve.

29 This flashed polymer entered a hot oil jacketed devolatilizer. Approximately 90 %
30 of the volatiles were removed from the polymer in the devolatilizer. The volatiles exit the
31 top of the devolatilizer. The remaining stream is condensed with a chilled water jacketed

1 exchanger and then enters a glycol jacket solvent / ethylene separation vessel. Solvent is
2 removed from the bottom of the vessel and ethylene vents from the top. The ethylene
3 stream is measured with a Micro-Motion™ mass flow meter. This measurement of
4 unreacted ethylene was used to calculate the ethylene conversion. The polymer separated
5 in the devolatilizer and was pumped out with a gear pump. The product is collected in
6 lined pans and dried in a vacuum oven at 140°C for 24 hr. Table 2 summarizes the
7 polymerization conditions and Table 3 the properties of the resulting polymers.
8

1 Table 2 - Ethylene/ α -Olefin Interpolymer Preparation Conditions*

Polymer	Reactor Temp °C	Solvent Flow lb/hr	Ethylene Flow lb/hr	Octene Flow lb/hr	Propylene Flow lb/hr	Hydrogen Flow sccm	C2 Conversion (%)	B/Ti Molar Ratio	MMAO/Ti Molar Ratio
1	150	25.4	2.65	1.37	-	143.8	90.10	1.16	4.99
2	150	25.2	2.65	1.20	-	139.9	90.00	1.23	5.03
3	150	25.6	2.65	1.15	-	149.9	90.40	1.21	4.93
4	151	25.0	2.65	1.90	-	167.3	90.22	1.24	5.00
5	150	25.2	2.65	1.72	-	197.0	89.75	1.21	4.93
6	151	25.1	2.65	1.85	-	144.2	90.47	1.25	4.97
7	150	25.2	2.65	1.67	-	175.4	90.36	1.20	4.95
8	148	25.5	2.65	1.00	-	126.6	89.95	1.15	4.99
9	151	25.2	2.65	1.30	-	111.5	90.09	1.14	4.97
10	150	25.1	2.65	1.24	-	108.2	90.42	1.20	9.92
11	148	25.1	2.65	1.35	-	107.9	90.05	1.21	4.99
12	148	25.3	2.65	1.15	-	113.9	90.00	1.19	5.01
13	147	25.1	2.65	1.31	-	106.9	90.09	1.24	5.04
14	147	25.5	2.65	1.00	-	114.9	90.02	1.15	4.98
15	151	25.2	2.65	1.30	-	83.7	89.88	1.13	4.93
16	150.5	25.2	2.65	-	0.60	159.9	90.2	1.22	5.01
17	150.1	25.2	2.65	-	0.75	164.8	89.9	1.20	4.94
18	150.2	25.2	2.65	-	0.45	165.5	90.0	1.18	4.91
19	150.1	25.2	2.65	-	0.90	166.0	89.8	1.20	5.02
20	150.4	25.2	2.65	-	0.70	124.9	90.3	1.26	5.04
21	150.4	25.2	2.65	-	0.52	126.9	89.9	1.21	5.05
22	149.9	25.2	2.62	-	0.70	94.9	90.0	1.22	4.72
23	150.7	25.2	2.65	-	0.52	96.2	90.5	1.20	5.02

2 * The catalyst for all polymerizations was $(C_5Me_4SiMe_2N^tBu)Ti(\eta^4-1,3\text{-pentadiene})$ prepared according to Example 17 of
3 US Patent 5,556,928, the entire disclosure of which patent is incorporated herein by reference. The primary cocatalyst for
4 all polymerisations was Armeenium Borate [methylbis(hydrogenatedtallowalkyl) ammonium tetrakis (pentafluoro
5 phenyl) borate prepared as in U.S. Patent # 5,919,983, Ex. 2, the entire disclosure of which patent is incorporated herein
6 by reference. The secondary cocatalyst for all polymerizations was a modified methylaluminoxane (MMAO) available
7 from Akzo Nobel as MMAO-3A (CAS# 146905-79-10).

8

Table 3 - Properties of Ethylene/ α -Olefin Interpolymers

Polymer #	Viscosity @ 300 °F (cP)	Density (g/cm ³)	M _w	M _n	M _w /M _n	Wt% Com.	Mol% Com.	Drop Point (°C)	T _m 1 (°C)	T _m 2 (°C)	Heat of Fusion (J/g)	% Cryst	T _c 1 (°C)	T _c 2 (°C)
1	1,375	0.9073	8,890	3,420	2.60	19.2	5.6	105.6	99.1	103.3	122.6	42	90.0	50.5
2	1,442	0.9125	9,000	3,480	2.59	20	5.9	108.9	102.1	107.0	142.4	49	92.4	54.1
3	1,490	0.9153	8,940	3,380	2.64	15.7	4.4	109.5	104.0	107.6	140.2	48	94.6	55.9
4	1,638	0.8953	9,310	3,650	2.55	28.3	9.0	99.3	93.3	83.2	99.1	34	82.5	
5	1,716	0.8991	9,280	3,640	2.55	22.2	6.7	102.4	95.8		113.6	39	85.8	
6	2,229	0.8958	10,100	4,080	2.48	27.8	8.8	100.0	93.3	83.3	99.7	34	82.8	
7	2,253	0.9014	9,970	4,070	2.45	21.9	6.5	103.6	96.5		111.9	38	86.1	
8	2,959	0.9146	10,800	4,860	2.22	16.4	4.7	110.5	103.3	106.9	134.9	46	93.1	53.9
9	3,054	0.9085	10,700	4,080	2.62	19.1	5.6	107.4	101.1		126.4	43	91.4	51.1
10	3,029	0.9092	10,800	4,310	2.51	18.2	5.3	107.1	99.7		122.3	42	89.1	48.9
11	3,139	0.9035	11,000	4,850	2.27	20.6	6.1	104.5	97.4		112.1	38	86.9	
12	3,545	0.9088	11,600	5,370	2.16	18.7	5.4	107.4	100.3		123.8	42	88.9	47.9
13	3,802	0.9039	11,700	5,150	2.27	20.5	6.1	105.1	97.4		114.3	39	85.9	
14	4,109	0.9143	11,800	5,300	2.23	16.8	4.8	110.7	103.5		135.3	46	93.3	53.2
15	5,899	0.9078	12,800	4,890	2.62	19.3	5.6	107.5	100.4		123.8	42	90.2	48.5
16	1,458	0.9152	9,070	4,030	2.25	12.2	8.5	103.1	85.6	99.5	147.2	50	87.7	58.1
17	1,503	0.9088	9,010	3,920	2.30	14.9	10.5	99.2	79.7	95.0	128.4	44	82.9	53.1
18	1,545	0.9267	9,100	3,900	2.33	11.3	7.8	110.2	106.4		163.9	56	95.5	
19	1,557	0.9003	9,060	3,970	2.28	18.9	13.4	94.6	74.9	90.2	112.4	38	77.9	68.6
20	2,884	0.9172	10,700	4,550	2.35	11.3	7.8	105.5	101.2		135.6	46	89.9	59.1
21	3,005	0.9187	10,800	4,690	2.30	11.1	7.7	106.0	102.1		149.3	51	90.7	60.4
22	5,889	0.9161	12,900	5,460	2.36	11.8	8.2	96.7			143.1	49.0	91	59.2
23	6,019	0.9162	12,900	5,470	2.36	11.9	8.3	106.2	87.5	100.5	141.1	48	89.4	58.5

1 Example 2. Preparation of Adhesive Formulations with Tackifier.

2 Ingredients were blended in a metal container to a total weight of 100g. Tackifier
3 resin was added into the container and allowed to heat for 10 minutes with a heating
4 mantle for temperature control. The polymer was slowly added over 3-5 minutes. Once
5 melted, the ingredients were mixed by hand using a metal spatula at a moderate rate of
6 speed. After complete addition of the polymer, the adhesive was allowed to mix an
7 additional 15 minutes to assure uniformity. The final adhesive temperature in all cases
8 was 350-360° F. A single tackifier was used in some formulations, while other
9 formulations used a combination of tackifiers.

10
11 Example 3. Evaluation of Adhesive Formulations

12 The adhesive formulations prepared according to Example 2 were evaluated for
13 their adhesive properties using the testing methods previously described. The properties
14 of these adhesive formulations are summarized in Tables 4 -6, and are compared with the
15 properties of several commercially available adhesives (Table 7).

16 Most of the ethylene/alpha-olefin polymers synthesized using ethylene and 1-
17 octene showed good performance when fiber tear was evaluated over the higher range of
18 temperatures (between 77 degrees F and 140 degrees F). Several of these formulations
19 also were effective at 35 degrees F.
20

Table 4 - Properties of Hot Melt Adhesives of the Present Invention (Ethylene/Octene Interpolymer)

Ex #	Polymer Sample #	Polymer (wt%)	Escorez 5637 (wt%)	FiberTear (%)					PAFT (°F)	SAFT (°F)	Viscosity @ 350 °F (cP)
				0 °F	35 °F	77 °F	120 °F	140 °F			
1	1	85.0	15.0	100	100	100	0	0	86	202	680
2	1	72.5	27.5	0	0	100	100	100	118	198	510
3	1	60.0	40.0	0	0	0	100	100	136	191	430
4	3	85.0	15.0	0	0	0	0	0	108	212	620
5	3	72.5	27.5	0	0	0	100	100	122	207	525
6	3	60.0	40.0	0	0	0	100	100	142	203	465
7	4	85.0	15.0	100	100	100	0	0	93	183	680
8	4	72.5	27.5	0	100	100	100	100	100	178	540
9	4	60.0	40.0	0	0	100	100	100	126	174	440
10	6	85.0	15.0	100	100	100	0	0	86	186	910
11	6	72.5	27.5	50	100	100	0	0	115	181	790
12	6	60.0	40.0	0	0	100	100	100	128	176	585
13	7	60.0	40.0	0	0	0	100	100	135	185	750
14	8	78.0	22.0	0	0	100	100	100	116		1,120
15	8	73.0	27.0	0	0	100	100	50	127		1,000
16	8	68.0	32.0	0	0	100	100	0	140		930
17	9	72.5	27.5	0	100	100	100	100	122	201	1,020
18	10	78.0	22.0	0	0	100	100	100	110	205	1,115
19	10	73.0	27.0	0	0	100	100	100	119	203	1,050
20	10	68.0	32.0	0	0	100	100	100	128	201	950
21	11	78.0	22.0	0	100	100	100	100	106		1,240
22	11	73.0	27.0	0	100	100	100	100	124		1,090
23	11	68.0	32.0	0	0	100	100	100	127		985
24	12	78.0	22.0	0	50	100	100	100	112		1,290
25	12	73.0	27.0	0	0	100	100	100	125		1,190
26	12	68.0	32.0	0	0	100	100	100	130		1,150
27	13	78.0	22.0	0	100	100	50	100	111		1,320
28	13	73.0	27.0	0	100	100	100	100	119		1,310
29	13	68.0	32.0	0	0	100	100	100	131		1,260
30	14	78.0	22.0	0	0	100	100	50	123		1,380
31	14	73.0	27.0	0	0	100	50	50	129		1,340
32	14	68.0	32.0	0	0	100	50	50	136		1,230
33	15	85.0	15.0	100	100	100	100	100	112	206	2,250
34	15	72.5	27.5	0	100	100	100	100	126	203	1,820
35	15	60.0	40.0	0	0	0	100	100	142	199	1,435

Table 5 - Properties of Hot Melt Adhesives of the Present Invention (Ethylene/Propylene Interpolymer)

Ex #	Polymer Sample # (wt%)	Tackifier* (wt%)	FiberTear (%)					PAFT (°F)	SAFT (°F)	Viscosity @ 350 °F (cP)
			0 °F	35 °F	77 °F	120 °F	140 °F			
36	16	15.0	0	0	0	0	0	92	201	745
37	16	27.5	0	0	0	100	100	106	199	590
38	18	15.0	0	0	0	0	0	108	215	800
39	18	40.0	0	0	0	0	0	138	207	490
40	19	15.0	0	100	100	0	0	93	176	695
41	19	27.5	0	0	100	0	0	92	171	630
42	19	40.0	0	0	0	100	100	123	168	485
43	21	15.0	0	100	0	0	0	111	206	1370
44	21	40.0	0	0	0	0	0	139	197	822
45	22	15.0	0	100	0	0	0	108	206	2490
46	22	27.5	0	0	0	0	100	128	203	1975
47	22	40.0	0	0	0	0	0	143	198	1490

*In all examples the tackifier used was Escorez 5637

Table 6 - Properties of Hot Melt Adhesives of the Present Invention (mixed tackifiers)

Ex #	Polymer Sample # (wt%)	Tackifier (wt%)		FiberTear (%)				PAFT (°F)	SAFT (°F)	Viscosity (cP)	
		E-5400 ¹	E-5637 ²	0 °F	35 °F	77 °F	120 °F			300 °F	350 °F
48	4 (70.0)	22.5	7.5	10	100	-	20	98	177	1090	550
49	4 (65.0)	17.5	17.5	0	80	-	80	118	174	1200	520
50	4 (75.0)	12.5	12.5	20	100	-	10	100	181	1180	610
51	4 (75.0)	0	25.0	50	100	-	100	104	182	1250	640
52	4 (65.0)	0	35.0	0	0	-	100	114	177	1100	545
53	4 (67.5)	7.5	25.0	0	80	-	100	112	177	1210	580
54	4 (70.0)	30.0	0	10	100	-	60	95	175	1050	535
55	4 (65.0)	0	35.0	0	0	-	100	119	176	1050	550
56	4 (75.0)	25.0	0	50	100	-	50	94	179	1145	595
57	4 (65.0)	35.0	0	0	100	-	100	100	173	992	500
58	4 (65.0)	35.0	0	10	100	-	100	101	174	990	490
59	4 (75.0)	0	25.0	0	100	-	100	104	179	1260	650
60	4 (70.0)	0	30.0	0	80	-	100	113	180	1185	600
61	6 (60.0)	40.0	0	0	0	-	100	127	177	1330	625
62	6 (67.5)	32.5	0	0	0	-	80	119	179	1490	720
63	6 (75.0)	25.0	0	0	100	-	20	102	186	1640	830
64	6 (60.0)	40.0	0	0	0	-	100	125	178	1340	630
65	6 (65.0)	35.0	0	0	0	-	80	122	178	1420	680
66	6 (75.0)	25.0	0	0	100	-	25	100	188	1630	825
67	6 (70.0)	30.0	0	0	50	-	100	118	181	1510	745

Table 7 - Properties of Commercial Hot Melt Adhesives of Prior Art

Comp Ex #	Name	Type	Viscosity @ 300 °F (cP)	Viscosity @ 350 °F (cP)	FiberTear					PAFT (°F)	SAFT (°F)
					0 °F	35 °F	77 °F	120 °F	140 °F		
1	ADVANTRA HL-9250	AFFINITY*-Based	1680	860	1.0	1.0	1.0	1.0	1.0	142	198
2	ADVANTRA HL-9256	AFFINITY*-Based	1560	750	0	1.0	1.0	1.0	1.0	151	192
	BAM Futura 1		1440	650	0	1.0	1.0	1.0	1.0	136	192
	EVA 1	EVA-Based	1587					1.0			150
3	Fuller HL-7268	EVA-Based		960			1.0	1.0	1.0	144	192
4	Fuller HL-2835	EVA-Based		1,070	1.0	1.0	1.0	1.0	1.0	126	153
5	Henkel 80-8488	EVA-Based		1,080		1.0	1.0	1.0	1.0	150	176
6	Henkel 80-8368	EVA-Based		970		1.0	1.0	1.0	1.0	142	190

* AFFINITY is a homogeneous polymer, which is a trademark of and available from The Dow Chemical Company.

1 These results show that combinations of these polymer and tackifier(s) can
2 produce an adhesive with properties that can be formulated to meet the needs of a wide
3 range of adhesive applications.

4 The results also show that these novel polymers, when formulated with a suitable
5 tackifier, have adhesive properties that are either equivalent to or better than a
6 conventional EVA hot melt adhesive which is formulated with wax and tackifier and
7 EVA resin. The results also demonstrate that the novel polymers of the present
8 invention, when compounded with select tackifiers, have properties comparable to a
9 premium hot melt adhesive as demonstrated by fiber tear.

10
11 Example 4. Thermal Stability Testing

12 Metallocene derived ethylene resins are known to be quite thermally stable when
13 compared to conventional EVA resins. To test the thermal stability of the present novel
14 resins a novel ethylene octene copolymer resin of the present invention with a density of
15 0.9032 g/cm³ were placed in a convection oven at 350 degrees F. The results, shown in
16 Table 8, illustrate the thermal stability of the novel polymer. Gardner color is a common
17 way to measure thermal degradation of hot melt adhesives. Color generation is directly
18 related to thermal degradation; therefore, the higher the Gardner number, the greater the
19 color generation in the polymer.

20
21 Table 8. Thermal stability of novel polymer

Time	Observation Gardner Color
0 hours	2
48 hours	+2, clear
96 hours	7, clear

1 Example 5. Preparation of Ethylene/ α -Olefin Interpolymers Using A Dual Metallocene
2 Catalyst System.

3
4 A series of ethylene/ α -olefin interpolymers were also prepared in a 1 gallon , oil
5 jacketed, Autoclave continuously stirred tank reactor (CSTR). A magnetically coupled
6 agitator with Lightning A-320 impellers provided the mixing. The reactor ran liquid full
7 at 475 psig (3,275 kPa). Process flow was in at the bottom and out of the top. A heat
8 transfer oil was circulated through the jacket of the reactor to remove some of the heat of
9 reaction. At the exit of the reactor was a Micro-MotionTM flow meter that measured flow
10 and solution density. All lines on the exit of the reactor were traced with 50 psi (344.7
11 kPa) steam and insulated.

12 ISOPAR-E solvent and comonomer were supplied to the reactor at 30 psig
13 pressure. The solvent feed to the reactors was measured by a Micro-MotionTM mass flow
14 meter. A variable speed diaphragm pump controlled the solvent flow rate and increased
15 the solvent pressure to reactor pressure. The comonomer was metered by a Micro-
16 MotionTM mass flow meter and flow controlled by a Research control valve. The
17 comonomer stream was mixed with the solvent stream at the suction of the solvent pump
18 and was pumped to the reactor with the solvent. The remaining solvent was combined
19 with ethylene and (optionally) hydrogen and delivered to the reactor. The ethylene stream
20 was measured by a Micro-MotionTM mass flow meter just prior to the Research valve
21 controlling flow. Three Brooks flow meter/controllers (1 - 200 sccm and 2 - 100sccm)
22 were used to deliver hydrogen into the ethylene stream at the outlet of the ethylene
23 control valve.

24 The ethylene or ethylene / hydrogen mixture combined with the solvent /
25 comonomer stream at ambient temperature. The temperature of the solvent/monomer as it
26 enters the reactor was controlled with two heat exchangers. This stream enters the
27 bottom of the 1 gallon CSTR.

28 In an inert atmosphere box, a solution of the transition metal compounds was
29 prepared by mixing the appropriate volumes of concentrated solutions of each of the two
30 components with solvent to provide the final catalyst solution of known concentration

1 and composition. This solution was transferred under nitrogen to a pressure vessel
2 attached to a high-pressure metering pump for transport to the polymerization reactor.

3 In the same inert atmosphere box, solutions of the primary cocatalyst,
4 methylbis(hydrogenatedtallowalkyl) ammonium tetrakis (pentafluoro phenyl) borate and
5 the secondary cocatalyst, MMAO Type 3A, were prepared in solvent and transferred to
6 separate pressure vessels as described for the catalyst solution. The ratio of Al to the
7 transition metal ("TM") and Boron to TM was established by controlling the volumetric
8 flow output if the individual metering pumps to attain the molar ratios in the
9 polymerization reactor as presented in Table 9. The three component catalyst system and
10 its solvent flush also enter the reactor at the bottom but through a different port than the
11 monomer stream.

12 Polymerization was stopped with the addition of catalyst kill into the reactor
13 product line after the meter measuring the solution density. Other polymer additives
14 could be added with the catalyst kill. The reactor effluent stream then entered a post
15 reactor heater that provides additional energy for the solvent removal flash. This flash
16 occurs as the effluent exits the post reactor heater and the pressure is dropped from 475
17 psig down to 10 at the reactor pressure control valve.

18 This flashed polymer entered a hot oil jacketed devolatilizer. Approximately 90 %
19 of the volatiles were removed from the polymer in the devolatilizer. The volatiles exit the
20 top of the devolatilizer. The remaining stream is condensed with a chilled water jacketed
21 exchanger and then enters a glycol jacket solvent / ethylene separation vessel. Solvent is
22 removed from the bottom of the vessel and ethylene vents from the top. The ethylene
23 stream is measured with a Micro-Motion™ mass flow meter. This measurement of
24 unreacted ethylene was used to calculate the ethylene conversion. The polymer separated
25 in the devolatilizer and was pumped out with a gear pump. The product is collected in
26 lined pans and dried in a vacuum oven at 140°C for 24 hr.

27 Additives (for example, antioxidants, pigments, etc.) could be incorporated into
28 the interpolymer products and all polymers could be stabilized with approximately 1000
29 ppm IRGANOX® 1010 and 2000 ppm IRGAFOS 168. Both IRGANOX® and
30 IRGAFOS™ are made by and trademarks of Ciba Geigy Corporation. IRGAFOS™ 168
31 is a phosphite stabilizer and IRGANOX® 1010 is a hindered polyphenol stabilizer (e.g.,

1 tetrakis [methylene 3-(3,5-di *t*-butyl-4-hydroxyphenylpropionate)]-methane. Table 9
 2 summarizes the polymerization conditions and Table 10 the properties of the resulting
 3 polymers.

6 Table 9 - Ethylene/ α -Olefin Interpolymer Preparation Conditions*

Ex	Reactor Temp °C	Solvent Flow lb/hr	Ethylene Flow lb/hr	Octene Flow lb/hr	Hydrogen Flow sccm	C2 Conversion (%)	B ^a /Tr Molar Ratio	MMAO ^b /Tr ^c Molar Ratio	Catalyst	Mole Ratio Catalyst
1	150.32	25.20	2.68	1.25	174.48	89.47	1.21	10.07	CATS-1/2	1:1
2	150.50	25.76	2.65	0.86	111.75	89.69	1.47	6.01	CATS-1/2	1:3
3	150.38	25.80	2.65	0.76	113.80	90.37	1.51	6.04	CATS-1/2	1:3
4	149.88	25.77	2.65	0.85	150.35	80.15	1.37	5.96	CATS 1/2	1:3
5	129.73	20.87	2.65	1.03	97.77	90.46	1.47	5.99	CATS 3/1	1:1
6	130.03	20.81	2.65	1.06	69.90	90.13	1.48	5.83	CATS 3/1	20:1
7	119.13	20.78	2.65	1.17	47.98	90.03	1.49	5.93	CATS 3/1	20:1
8	149.65	25.51	2.65	1.00	83.20	90.40	1.06	4.95	CATS-4/1	1:1
9	120.28	25.20	2.65	1.60	13.45	90.44	1.08	4.91	CATS 3/1	10:1
10	150.20	25.60	2.65	0.73	121.97	90.35	1.08	4.95	CATS 4/2	1:2

7 ^aThe primary cocatalyst for all polymerisations was Armeenium Borate [methylbis(hydrogenatedtallowalkyl)
 8 ammonium tetrakis (pentafluoro phenyl) borate prepared as in U.S. Patent # 5,919,983, Ex. 2, the entire disclosure of
 9 which patent is incorporated herein by reference.

10 ^bThe secondary cocatalyst for all polymerizations was a modified methylaluminoxane (MMAO) available from Akzo
 11 Nobel as MMAO-3A (CAS# 146905-79-10).

12 ^cFor Examples 1-4, 8 and 10 in Table 11 the term Tr refers to the total titanium content of the mixed catalyst system.
 13 For runs 5-7 and 9 the term Tr refers to the Zr content only of the mixed catalyst system.

14 ^dCAT 1 was (C₅Me₄SiMe₂N^tBu)Ti(η^4 -1,3-pentadiene) prepared according to Example 17 of US Patent 5,556,928,
 15 the entire disclosure of which patent is incorporated herein by reference. CAT 2 was (1H-cyclopenta[1]-
 16 phenanthrene-2-yl)dimethyl (t-butylamido) silanetitanium dimethyl prepared according to Examples 1 and 2 of US
 17 Patent 5,150,297, the entire disclosure of which patent is incorporated herein by reference. CAT 3 was
 18 (C₅Me₄SiMe₂N^tBu)ZrMe₂ Prepared according to Examples 1 and 86 of US Patent 5,703,187, the entire disclosure of
 19 which patent is incorporated herein by reference. CAT 4 was [N-(1,1-dimethylethyl)-1,1-dimethyl-1-[1,2,3,4,5- η]-
 20 3,4-diphenyl-2,4-cyclopentadienyl-1-yl]silanaminato(2)- κ N]-dimethyl-titanium, prepared according to Examples 1
 21 and 2 of WO 02/092610, the entire disclosure of which patent is incorporated herein by reference.

Table 10 - Properties of Ethylene/ α -Octene Interpolymers

Polymer #	Viscosity @ 300 °F (cP)	Density (g/cm ³)	M _w	M _n	M _w /M _n	Wt% Com.	Mol% Com.	Drop Point (°C)	T _m 1 (°C)	T _m 2 (°C)	Heat of Fusion (J/g)	% Cryst	T _c 1 (°C)	T _c 2 (°C)
1	1,600	0.8941	9,570	4,180	2.29	23.40	7.10	113.3	81.2	107.0	96.2	33	97.1	55.0
2	2,879	0.9040	11,200	5,030	2.23	19.80	5.81	116.9	86.3	110.3	113.3	39	99.8	73.4
3	2,859	0.9083	11,300	5,220	2.16	18.30	5.30	117.8	89.4	111.4	121.4	42	101.2	77.1
4	2,744	0.9092	10,900	5,060	2.15	18.10	5.23	118.4	90.0	112.3	125.9	43	102.7	78.2
5	2,804	0.9091	11,200	2,700	4.15	18.40	5.34	109.6	103.3		120.7	41	91.1	52.1
6	2,889	0.9089	12,000	2,080	5.77	18.90	5.50	112.1	95.1	107.2	125.8	43	94.7	
7	2,684	0.9052	12,800	1,590	8.05	19.30	5.64	113.5	93.7	110.2	130.9	45	97.1	81.1
8	3,047	0.9086	11,000	4,610	2.39	17.7	5.1	109.6	96.7	103.3	130.2	45	93.6	54.2
9	3,113	0.9067	17,000	1,130	15.04	18.8	5.5	116.1	93.1	113.7	136.7	47	100.8	
10	2,855	0.9084	10,800	3,940	2.74	18.1	6.3	114.6	93.3	105.6	134.7	46	95.0	82.3

1 Example 6. Preparation of Adhesive Formulations with Tackifier, Using Ethylene/ α -
2 Olefin Interpolymers of Example 5.

3
4 As has been described previously in Example 2, ingredients for the hot melt
5 adhesive compositions were blended in a metal container to a total weight of 100g.
6 Tackifier resin was added into the container and allowed to heat for 10 minutes with a
7 heating mantle for temperature control. The polymer was slowly added over 3-5 minutes.
8 Once melted, the ingredients were mixed by hand using a metal spatula at a moderate rate
9 of speed. After complete addition of the polymer, the adhesive was allowed to mix an
10 additional 15 minutes to assure uniformity. The final adhesive temperature in all cases
11 was 350-360° F.

12
13 Example 7. Evaluation of Adhesive Formulations.

14 The properties of the adhesive formulations of Example 6 are summarized in
15 Table 11 and compared with the properties of the commercially available adhesives
16 summarized in Table 7.

Table 11. Properties of Hot Melt Adhesives Made From Ethylene/Octene Interpolymer of the Present Invention

Polymer Ex #	Polymer (wt%)	Escorez 5637 (wt%)	Fiber Tear (%)					PAFT (°F)	SAFT (°F)	Viscosity @ 350 °F (cP)
			0 °F	35 °F	77 °F	120 °F	140 °F			
1	78	22	0	25		100	100	110	205	1,115
1	73	27	0	0		100	100	119	203	1,050
1	68	32	0	0		100	100	128	201	950
2	78	22	0	100	100	100	75	110	211	1,060
2	73	27	0	100	100	100	100	118	208	935
2	68	32	0	0	100	100	100	131	208	820
3	78	22	0	50	100	100	100	110	215	1,080
3	73	27	0	25	100	100	100	132	212	980
3	68	32	0	0	100	100	100	156	211	660
4	78	22	0	50	100	100	75	120	215	570
4	73	27	0	25	100	100	100	122	213	500
4	68	32	0	0	100	100	100	132	211	470
5	78	22	0	100	100	100	50	111	203	1,050
5	73	27	0	25	100	100	100	115	202	960
5	68	32	0	0	100	100	100	118	200	860
6	78	22	10	50	100	100	100	104	203	1,000
6	73	27	0	10	100	100	100	115	202	945
6	68	32	0	0	100	100	100	124	200	850
7	78	22	25	25	NM*	50	50	95	209	925
7	73	27	0	25	100	100	75	109	207	840
7	68	32	0	0	100	100	100	127	205	755
8	83	17	0	100	NM	100	100	90	214	1300
8	78	22	0	50	NM	100	100	109	208	1205
8	73	27	0	0	NM	100	100	126	207	1100
8	68	32	0	0	NM	100	100	128	207	1035
9	83	17	100	100	NM	50	0	90	212	1140
9	78	22	100	100	NM	100	0	90	210	1070
9	73	27	75	100	NM	100	75	90	208	930
9	68	32	0	100	NM	100	100	111	208	810
10	83	17	0	100	NM	100	100	105	205	1175
10	78	22	0	100	NM	100	100	112	204	1115
10	73	27	0	0	NM	100	100	126	202	1040
10	68	32	0	0	NM	100	100	131	203	920

* NM = not measured

1 CLAIMS.

2
3 We claim:4
5 1. A hot melt adhesive composition consisting essentially of:6 A) from about 40 to 100 percent by weight (based on the final weight of the hot
7 melt adhesive composition) of a homogenous ethylene/ α -olefin interpolpolymer; and8 B) from 0 to about 60 percent by weight (based on the final weight of the hot melt
9 adhesive composition) of one or more tackifiers.10
11 2. The hot melt adhesive composition of Claim 1 wherein;12 A) the homogenous ethylene/ α -olefin interpolpolymer is present in an amount of
13 from about 60 to about 85 percent by weight (based on the final weight of the hot melt
14 adhesive composition) and the homogenous ethylene/ α -olefin interpolpolymer is
15 characterized by having:16 i) a density of from about 0.880 to about 0.930 g/cm³;17 ii) a number average molecular weight (Mn) of from about 1,000 to about
18 9,000; and19 iii) a Brookfield Viscosity (measured at 300°F) of from about 500 to about
20 7,000 cP and21 B) the one or more tackifiers is present in an amount of from about 15 to about 40
22 percent by weight (based on the final weight of the hot melt adhesive composition); and
23 wherein

24 C) the hot melt adhesive composition is characterized by having:

25 i) a Brookfield Viscosity (measured at 350°F) of from about 400 to about
26 2,000 cP;27 ii) a Peel Adhesion Failure Temperature ("PAFT") of greater than or equal
28 to 110°F; and29 iii) a Shear Adhesion Failure Temperature ("SAFT") of greater than or
30 equal to 140°F.

31

1 3. The hot melt adhesive composition of Claim 1 wherein;

2 A) the homogenous ethylene/ α -olefin interpolymer is present in an amount of
3 from about 65 to about 80 percent by weight (based on the final weight of the hot melt
4 adhesive composition) and the homogenous ethylene/ α -olefin interpolymer is
5 characterized by having:

- 6 i) a density of from about 0.890 to about 0.920 g/cm³;
- 7 ii) a number average molecular weight (Mn) of from about 1,250 to about
8 7,000; and
- 9 iii) a Brookfield Viscosity (measured at 300°F) of from about 1,000 to
10 about 6,000 cP; and

11 B) the one or more tackifiers is present in an amount of from about 20 to about 35
12 percent by weight (based on the final weight of the hot melt adhesive composition); and
13 wherein

14 C) the hot melt adhesive composition is characterized by:

- 15 i) having a Brookfield Viscosity (measured at 350°F) of from about 500
16 to about 1,400 cP;
- 17 ii) having a Peel Adhesion Failure Temperature ("PAFT") of greater than
18 or equal to 115°F;
- 19 iii) having a Shear Adhesion Failure Temperature ("SAFT") of greater
20 than or equal to 150°F; and
- 21 iv) exhibiting 100% paper tear at 35°F - 140°F.

22

23 4. The hot melt adhesive composition of Claim 1 wherein;

24 A) the homogenous ethylene/ α -olefin interpolymer is characterized by having:

- 25 i) a density of from about 0.895 to about 0.915 g/cm³;
- 26 ii) a number average molecular weight (Mn) of from about 1,500 to about
27 6,000; and
- 28 iii) a Brookfield Viscosity (measured at 300°F) of from about 1,500 to
29 about 5,000 cP; and

30 B) the hot melt adhesive composition is characterized by:

- 1 i) having a Brookfield Viscosity (measured at 350°F) of from about 750 to
- 2 about 1,200 cP;
- 3 ii) having a Peel Adhesion Failure Temperature ("PAFT") of greater than
- 4 or equal to 120°F;
- 5 iii) having a Shear Adhesion Failure Temperature ("SAFT") of greater
- 6 than or equal to 170°F; and
- 7 iv) exhibits 100% paper tear at 0°F - 140°F.

8

9 5. The hot melt adhesive composition of Claim 1 wherein;

10 A) the homogenous ethylene/ α -olefin interpolymer is characterized by having:

- 11 i) a density of from about 0.893 to about 0.930 g/cm³;
- 12 ii) a number average molecular weight (Mn) of from about 1000 to about
- 13 6,000; and
- 14 iii) a Brookfield Viscosity (measured at 300°F) of from about 1,500 to
- 15 about 5,000 cP; and

16 B) the hot melt adhesive composition is characterized by:

- 17 i) having a Brookfield Viscosity (measured at 350°F) of from about 400 to
- 18 about 1,400 cP;
- 19 ii) having a Peel Adhesion Failure Temperature ("PAFT") of greater than
- 20 or equal to 90°F;
- 21 iii) having a Shear Adhesion Failure Temperature ("SAFT") of greater
- 22 than or equal to 200°F.; and
- 23 v) exhibits 100% paper tear at 140°F.

24

25 6. The hot melt adhesive composition of Claim 5 wherein;

26 A) the homogenous ethylene/ α -olefin interpolymer is characterized by having:

- 27 i) a density of from about 0.894 to about 0.910 g/cm³;
- 28 ii) a number average molecular weight (Mn) of from about 1100 to about
- 29 5,300; and

1 iii) a Brookfield Viscosity (measured at 300°F) of from about 1,600 to
2 about 3,200 cP; and

3 B) the hot melt adhesive composition is characterized by:

4 i) having a Brookfield Viscosity (measured at 350°F) of from about
5 700 to about 1,200 cP;

6 ii) having a Peel Adhesion Failure Temperature ("PAFT") of greater than
7 or equal to 90°F;

8 iii) having a Shear Adhesion Failure Temperature ("SAFT") of greater
9 than or equal to 200°F.; and

10 iv) exhibits 100% paper tear at 140°F.

11

12 7. The hot melt adhesive composition of any of Claims 2,3,4, or 6 wherein;

13 A) the homogenous ethylene/ α -olefin interpolymer is an interpolymer of ethylene
14 and one or more C₃-C₃₀ α -olefins; and

15 B) the one or more tackifiers is selected from the group consisting of aliphatic
16 hydrocarbon resins, hydrogenated hydrocarbon resins, C₅ aliphatic or aromatic
17 hydrocarbon resins or an aromatically modified C₅ aliphatic or aromatic hydrocarbon
18 resins and combinations thereof; and

19 wherein the adhesive composition is capable of binding a fibrous cellulosic article
20 to an article selected from the group consisting of a fibrous cellulosic article, wood,
21 metal, glass, plastic, and combinations thereof.

22

23 8. The hot melt adhesive composition of Claims 4 or 5 wherein;

24 A) in the homogenous ethylene/ α -olefin interpolymer, the α -olefin is selected
25 from the group consisting of from C₈ (1-Octene); C₁₀ (1-Decene), C₁₂ (1-dodecene), C₁₄
26 (1-duodecene), C₁₄ (1-tetradecene), C₁₆ (1-hexadecene), C₁₈ (1-octadecene), C₂₀₋₂₄⁺, C₂₄-
27 ₂₈ and C₃₀ and combinations thereof; and

28 B) the one or more tackifiers is characterized by having an acid number between 0
29 to and about 25.8.

30

9. The hot melt adhesive composition of Claim 7 or 8, further comprising one or more compounds chosen from the group consisting of stabilizers, plasticizers, fillers, antioxidants, preservatives, synergists, dyes, and pigments.

10. The hot melt adhesive composition of Claim 8, wherein the α -olefin is selected from the group consisting of 1-octene and propylene.

11. The hot melt adhesive composition of Claim 8 or 10, wherein the copolymer has a molecular weight distribution (Mw/Mn) ranging from about 2.1 – about 16.

12. The hot melt adhesive composition of Claim 8, 10 or 11, wherein the tackifier is present in an amount of from about 15 to about 35 percent by weight (based on the final weight of the hot melt adhesive composition).

13. A cellulosic article formed using a hot melt adhesive composition, the adhesive composition consisting essentially of;

A) from about 40 to 100 percent by weight (based on the final weight of the hot melt adhesive composition) of a homogenous ethylene/ α -olefin interpolymers; and

B) from 0 to about 60 percent by weight (based on the final weight of the hot melt adhesive composition) of one or more tackifiers.

14. The cellulosic article of Claim 13 wherein in the hot melt adhesive composition;

A) the homogenous ethylene/ α -olefin interpolymers is present in an amount of from about 60 to about 85 percent by weight (based on the final weight of the hot melt adhesive composition) and the homogenous ethylene/ α -olefin interpolymers is characterized by having:

i) a density of from about 0.880 to about 0.930 g/cm³;

ii) a number average molecular weight (Mn) of from about 1,000 to about 9,000; and

iii) a Brookfield Viscosity (measured at 300°F) of from about 500 to about 7,000 cP and

1 B) the one or more tackifiers is present in an amount of from about 15 to about 40
2 percent by weight (based on the final weight of the hot melt adhesive composition); and
3 wherein

4 C) the hot melt adhesive composition is characterized by having:

5 i) a Brookfield Viscosity (measured at 350°F) of from about 400 to about
6 2,000 cP;

7 ii) a Peel Adhesion Failure Temperature ("PAFT") of greater than or equal
8 to 110°F; and

9 iii) a Shear Adhesion Failure Temperature ("SAFT") of greater than or
10 equal to 140°F.
11

12 15. The cellulosic article of Claim 13 wherein in the hot melt adhesive composition;

13 A) the homogenous ethylene/ α -olefin interpolymers is present in an amount of
14 from about 65 to about 80 percent by weight (based on the final weight of the hot melt
15 adhesive composition) and the homogenous ethylene/ α -olefin interpolymers is
16 characterized by having:

17 i) a density of from about 0.890 to about 0.920 g/cm³;

18 ii) a number average molecular weight (Mn) of from about 1,250 to about
19 7,000; and

20 iii) a Brookfield Viscosity (measured at 300°F) of from about 1,000 to
21 about 6,000 cP; and

22 B) the one or more tackifiers is present in an amount of from about 20 to about 35
23 percent by weight (based on the final weight of the hot melt adhesive composition); and
24 wherein
25

26 C) the hot melt adhesive composition is characterized by:

27 i) having a Brookfield Viscosity (measured at 350°F) of from about 500
28 to about 1,400 cP;

29 ii) having a Peel Adhesion Failure Temperature ("PAFT") of greater than
30 or equal to 115°F;

- 1 iii) having a Shear Adhesion Failure Temperature ("SAFT") of greater
2 than or equal to 150°F; and
3 iv) exhibiting 100% paper tear at 140°F.
4

5 16. The cellulosic article of Claim 13 wherein in the hot melt adhesive composition;

6 A) the homogenous ethylene/ α -olefin interpolymer is characterized by having:

- 7 i) a density of from about 0.893 to about 0.930 g/cm³;
8 ii) a number average molecular weight (Mn) of from about 1,000 to about
9 6,000; and
10 iii) a Brookfield Viscosity (measured at 300°F) of from about 1,500 to
11 about 5,000 cP; and

12 B) the hot melt adhesive composition is characterized by:

- 13 i) having a Brookfield Viscosity (measured at 350°F) of from about 400 to
14 about 1,400 cP;
15 ii) having a Peel Adhesion Failure Temperature ("PAFT") of greater than
16 or equal to 90°F;
17 iii) having a Shear Adhesion Failure Temperature ("SAFT") of greater
18 than or equal to 200°F.; and
19 iv) exhibits 100% paper tear at 120°F.
20

21 17. The cellulosic article of Claim 13 wherein in the hot melt adhesive composition;

22 A) the homogenous ethylene/ α -olefin interpolymer is characterized by having:

- 23 i) a density of from about 0.894 to about 0.910 g/cm³;
24 ii) a number average molecular weight (Mn) of from about 1,100 to about
25 5,300; and
26 iii) a Brookfield Viscosity (measured at 300°F) of from about 1,600 to
27 about 3,200 cP; and

28 B) the hot melt adhesive composition is characterized by:

- 29 i) having a Brookfield Viscosity (measured at 350°F) of from about 700 to
30 about 1,200 cP;

ii) having a Peel Adhesion Failure Temperature ("PAFT") of greater than or equal to 90°F;

iii) having a Shear Adhesion Failure Temperature ("SAFT") of greater than or equal to 200°F.; and

iv) exhibits 100% paper tear at 140°F.

18. The cellulosic article of Claims 16 or 17, wherein in the hot melt adhesive composition:

A) the homogenous ethylene/ α -olefin interpolymer is an interpolymer of ethylene and one or more C₃-C₃₀ α -olefins; and

B) the one or more tackifiers is selected from the group consisting of aliphatic hydrocarbon resins, hydrogenated hydrocarbon resins, C₅ aliphatic or aromatic hydrocarbon resins or an aromatically modified C₅ aliphatic or aromatic hydrocarbon resins and combinations thereof.

19. The cellulosic article of Claim 18, wherein in the hot melt adhesive composition;

A) in the homogenous ethylene/ α -olefin interpolymer, the α -olefin is selected from the group consisting of from C₈ (1-Octene); C₁₀ (1-Decene), C₁₂ (1-dodecene), C₁₄ (1-duodecene), C₁₄ (1-tetradecene), C₁₆ (1-hexadecene), C₁₈ (1-octadecene), C₂₀₋₂₄⁺, C₂₄₋₂₈ and C₃₀ and combinations thereof; and

B) the one or more tackifiers is characterized by having an acid number between 0 to and about 25.8.

20. The cellulosic article of Claims 19, wherein the cellulosic article is selected from the group consisting of corrugated cardboard, kraft paper, linerboard, and paper.

1 21. A method of producing a polymer composition, the method comprising the steps of:
2 admixing:

3 an amount of from about 60 to about 85 percent by weight (based on the final
4 weight of the polymer composition) of a homogeneous ethylene/ α -olefin interpolymer;
5 the homogenous ethylene/ α -olefin interpolymer characterized by having:

6 i) a density of from about 0.880 to about 0.930 g/cm³;

7 ii) a number average molecular weight (Mn) of from about 1,000 to about 9,000;

8 and

9 iii) a Brookfield Viscosity (measured at 300°F) of from about 500 to about 7,000

10 cP and

11 admixing an amount of from 15 to about 40 percent by weight (based on the final weight
12 of the polymer composition) of a tackifier; and wherein

13 the polymer composition is characterized by having:

14 i) a Brookfield Viscosity (measured at 350°F) of from about 400 to about
15 2,000 cP;

16 ii) a Peel Adhesion Failure Temperature ("PAFT") of greater than or equal
17 to 110°F; and

18 iii) a Shear Adhesion Failure Temperature ("SAFT") of greater than or
19 equal to 140°F.

20
21 22. A method of producing a polymer composition, wherein:

22 A) the homogenous ethylene/ α -olefin interpolymer is characterized by having:

23 i) a density of from about 0.893 to about 0.930 g/cm³;

24 ii) a number average molecular weight (Mn) of from about 1,000 to about
25 6,000; and

26 iii) a Brookfield Viscosity (measured at 300°F) of from about 1,500 to
27 about 5,000 cP; and

28 B) the polymer composition is characterized by:

29 i) having a Brookfield Viscosity (measured at 350°F) of from about 400 to
30 about 1,400 cP;

- 1 ii) having a Peel Adhesion Failure Temperature ("PAFT") of greater than
- 2 or equal to 90°F;
- 3 iii) having a Shear Adhesion Failure Temperature ("SAFT") of greater
- 4 than or equal to 200°F.; and
- 5 iv) exhibits 100% paper tear at 140°F.

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
2 December 2004 (02.12.2004)

PCT

(10) International Publication Number
WO 2004/104127 A3

(51) International Patent Classification⁷: **C09J 123/08**,
C08L 23/08

(21) International Application Number:
PCT/US2004/015697

(22) International Filing Date: 19 May 2004 (19.05.2004)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
60/471,318 19 May 2003 (19.05.2003) US
10/666,488 19 September 2003 (19.09.2003) US

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(81) Designated States (*unless otherwise indicated, for every
kind of national protection available*): AE, AG, AL, AM,
AT, AU, AZ, BA, BB, BG, BR, BW, BY, BZ, CA, CH, CN,

CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, EG, ES, FI,
GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE,
KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD,
MG, MK, MN, MW, MX, MZ, NA, NI, NO, NZ, OM, PG,
PH, PL, PT, RO, RU, SC, SD, SE, SG, SK, SL, SY, TJ, TM,
TN, TR, TT, TZ, UA, UG, UZ, VC, VN, YU, ZA, ZM, ZW.

(84) Designated States (*unless otherwise indicated, for every
kind of regional protection available*): ARIPO (BW, GH,
GM, KE, LS, MW, MZ, NA, SD, SL, SZ, TZ, UG, ZM,
ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM),
European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI,
FR, GB, GR, HU, IE, IT, LU, MC, NL, PL, PT, RO, SE,
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GQ, GW, ML, MR, NE, SN, TD, TG).

Declarations under Rule 4.17:

- *as to applicant's entitlement to apply for and be granted a
patent (Rule 4.17(ii)) for all designations*
- *as to the applicant's entitlement to claim the priority of the
earlier application (Rule 4.17(iii)) for all designations*

Published:

- *with international search report*

(88) Date of publication of the international search report:
2 March 2006

*For two-letter codes and other abbreviations, refer to the "Guid-
ance Notes on Codes and Abbreviations" appearing at the begin-
ning of each regular issue of the PCT Gazette.*

(54) Title: NOVEL MULTIFUNCTIONAL POLYMER FOR USE IN HOT MELT ADHESIVE APPLICATIONS

(57) Abstract: Adhesives comprising one or more tackifiers and an ethylene/alpha-olefin interpolymer produced using either a single metallocene or dual metallocene catalyst system were formulated, and have adhesive characteristics over a broad temperature range. The inventive hot melt adhesive compositions ("HMAs") have properties comparable to those of commercially available, three component HMAs comprising ethylene vinyl acetate polymers, tackifier and wax. HMA embodiments include those wherein the ethylene/alpha-olefin interpolymers comprise ethylene and either 1-octene or propylene as copolymers.



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INTERNATIONAL SEARCH REPORT

International Application No

US2004/015697

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C09J123/08 C08L23/08

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C09J C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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A	the whole document	2-12, 14-22
X	US 6 319 979 B1 (ALBRECHT STEVEN W ET AL) 20 November 2001 (2001-11-20)	1,13
Y	column 5, line 5 - column 6, line 22	1-22
X	US 6 221 448 B1 (CHANAK MICHELLE M ET AL) 24 April 2001 (2001-04-24) claim 1	1,13
Y	column 5, line 10 - column 5, line 28	1-22



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

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- *Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- *Z* document member of the same patent family

Date of the actual completion of the international search

21 September 2004

Date of mailing of the international search report

29/09/2004

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INTERNATIONAL SEARCH REPORT

Int Application No
PCT/US2004/015697

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(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
2 December 2004 (02.12.2004)

PCT

(10) International Publication Number
WO 2004/104127 A3

(51) International Patent Classification⁷: **C09J 123/08**,
C08L 23/08

(21) International Application Number:
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(22) International Filing Date: 19 May 2004 (19.05.2004)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
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10/666,488 19 September 2003 (19.09.2003) US

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(81) Designated States (*unless otherwise indicated, for every
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AT, AU, AZ, BA, BB, BG, BR, BW, BY, BZ, CA, CH, CN,
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GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE,
KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD,
MG, MK, MN, MW, MX, MZ, NA, NI, NO, NZ, OM, PG,
PH, PL, PT, RO, RU, SC, SD, SE, SG, SK, SL, SY, TJ, TM,
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ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM),
European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI,
FR, GB, GR, HU, IE, IT, LU, MC, NL, PL, PT, RO, SE,
SI, SK, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN,
GQ, GW, ML, MR, NE, SN, TD, TG).

Declarations under Rule 4.17:

- *as to applicant's entitlement to apply for and be granted a
patent (Rule 4.17(ii))*
- *as to the applicant's entitlement to claim the priority of the
earlier application (Rule 4.17(iii))*

Published:

- *with international search report*
- *with amended claims*

(88) Date of publication of the international search report:
2 March 2006

Date of publication of the amended claims: 20 April 2006

*For two-letter codes and other abbreviations, refer to the "Guid-
ance Notes on Codes and Abbreviations" appearing at the begin-
ning of each regular issue of the PCT Gazette.*

(54) Title: NOVEL MULTIFUNCTIONAL POLYMER FOR USE IN HOT MELT ADHESIVE APPLICATIONS

(57) Abstract: Adhesives comprising one or more tackifiers and an ethylene/alpha-olefin interpolymer produced using either a single metallocene or dual metallocene catalyst system were formulated, and have adhesive characteristics over a broad temperature range. The inventive hot melt adhesive compositions ("HMAs") have properties comparable to those of commercially available, three component HMAs comprising ethylene vinyl acetate polymers, tackifier and wax. HMA embodiments include those wherein the ethylene/alpha-olefin interpolymers comprise ethylene and either 1-octene or propylene as copolymers.



WO 2004/104127 A3

AMENDED CLAIMS

[received by the International Bureau on 24 November 2004 (24.11.04);
original claims 1-22, replaced by new claim 1-22]

1 CLAIMS.

2

3 1. A hot melt adhesive composition consisting essentially of:

4 A) from about 40 to 100 percent by weight (based on the final weight of
5 the hot melt adhesive composition) of a homogeneous ethylene/ α -olefin interpolymers,
6 the homogeneous ethylene/ α -olefin interpolymers being characterized by having a
7 number average molecular weight (Mn) of from about 1,000 to about 9,000; and

8 B) from 0 to about 60 percent by weight (based on the final weight of the
9 hot melt adhesive composition) of one or more tackifiers.

10

11 2. The hot melt adhesive composition of Claim 1 wherein:

12 A) the homogeneous ethylene/ α -olefin interpolymers is present in an
13 amount of from about 60 to about 85 percent by weight (based on the final weight
14 of the hot melt adhesive composition) and the homogeneous ethylene/ α -olefin
15 interpolymers is further characterized by having:

16 i) a density of from about 0.880 to about 0.930 g/cm³; and

17 ii) a Brookfield Viscosity (measured at 149°C) ((300°F)) of from
18 about 5 to about 70 grams/(cm.second) (about 500 to about 7,000 cP); and

19 B) the one or more tackifiers is present in an amount of from about 15 to
20 about 40 percent by weight (based on the final weight of the hot melt
21 adhesive composition); and wherein

22 C) the hot melt adhesive composition is characterized by having:

23 i) a Brookfield Viscosity (measured at 177°C) ((350°F)) of from
24 about 4 to about 20 grams/(cm.second) (about 400 to about 2,000
25 cP);

26 ii) a Peel Adhesion Failure Temperature ("PAFT") of greater than
27 or equal to 43°C (110°F); and

28 iii) a Shear Adhesion Failure Temperature ("SAFT") of greater
29 than or equal to 60°C (140°F).

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1 3. The hot melt adhesive composition of Claim 1 wherein:

2 A) the homogeneous ethylene/ α -olefin interpolymer is present in an
3 amount of from about 65 to about 80 percent by weight (based on the final weight
4 of the hot melt adhesive composition) and the homogeneous ethylene/ α -olefin
5 interpolymer is further characterized by having:

6 i) a density of from about 0.890 to about 0.920 g/cm³;

7 ii) a number average molecular weight (Mn) of from about 1,250 to
8 about 7,000; and

9 iii) a Brookfield Viscosity (measured at 149°C) ((300°F)) of from
10 about 10 to about 60 grams/(cm.second) (about 1,000 to about
11 6,000 cP); and

12 B) the one or more tackifiers is present in an amount of from about 20 to
13 about 35 percent by weight (based on the final weight of the hot melt
14 adhesive composition); and wherein

15 C) the hot melt adhesive composition is characterized by:

16 i) having a Brookfield Viscosity (measured at 177°C) ((350°F)) of
17 from about 5 to about 14 grams/(cm.second) (about 500 to about
18 1,400 cP);

19 ii) having a Peel Adhesion Failure Temperature ("PAFT") of
20 greater than or equal to 46°C (115°F);

21 iii) having a Shear Adhesion Failure Temperature ("SAFT") of
22 greater than or equal to 66°C (150°F); and

23 iv) exhibiting 100% paper tear at a temperature range of 2°C-
24 60°C (35°F - 140°F).

25

26 4. The hot melt adhesive composition of Claim 1, wherein:

27 A) the homogeneous ethylene/ α -olefin interpolymer is further
28 characterized by having:

29 i) a density of from about 0.895 to about 0.915 g/cm³;

30 ii) a number average molecular weight (Mn) of from about 1,500
31 to about 6,000; and

- 1 iii) a Brookfield Viscosity (measured at 149°C ((300°F)) of from
2 about 15 to about 50 grams/(cm.second) (about 1,500 to about
3 5,000 cP); and
4 B) the hot melt adhesive composition is characterized by:
5 i) having a Brookfield Viscosity (measured at 177°C ((350°F)) of
6 from about 7.5 to about 12 grams/(cm.second) (about 750 to about
7 1,200 cP);
8 ii) having a Peel Adhesion Failure Temperature ("PAFT") of
9 greater than or equal to 49°C (120°F);
10 iii) having a Shear Adhesion Failure Temperature ("SAFT") of
11 greater than or equal to 77°C (170°F); and
12 iv) exhibits 100% paper tear at a temperature range of minus18°C
13 to 60°C (0°F - 140°F).
14
15 5. The hot melt adhesive composition of Claim 1 wherein:
16 A) the homogeneous ethylene/ α -olefin interpolymer is further
17 characterized by having:
18 i) a density of from about 0.893 to about 0.930 g/cm³;
19 ii) a number average molecular weight (Mn) of from about 1000 to
20 about 6,000; and
21 iii) a Brookfield Viscosity (measured at 149°C ((300°F)) of from
22 about 15 to about 50 grams/(cm.second) (about 1,500 to about
23 5,000 cP); and
24 B) the hot melt adhesive composition is characterized by:
25 i) having a Brookfield Viscosity (measured at 177°C ((350°F)) of
26 from about 4 to about 14 grams/(cm.second) (about 400 to about
27 1,400 cP);
28 ii) having a Peel Adhesion Failure Temperature ("PAFT") of
29 greater than or equal to 32°C (90°F);
30 iii) having a Shear Adhesion Failure Temperature ("SAFT") of
31 greater than or equal to 93°C. (200°F); and

- 1 iv) exhibits 100% paper tear at 60°C (140°F).
- 2
- 3 6. The hot melt adhesive composition of Claim 5 wherein;
- 4 A) the homogeneous ethylene/ α -olefin interpolymers is characterized by
- 5 having:
- 6 i) a density of from about 0.894 to about 0.910 g/cm³;
- 7 ii) a number average molecular weight (Mn) of from about 1100 to
- 8 about 5,300; and
- 9 iii) a Brookfield Viscosity (measured at 149°C ((300°F)) of from
- 10 about 16 to about 32 grams/(cm.second) (about 1,600 to about
- 11 3,200 cP); and
- 12 B) the hot melt adhesive composition is characterized by:
- 13 i) having a Brookfield Viscosity (measured at 177°C ((350°F)) of
- 14 from about 7 to about 12 grams/(cm.second) (about 700 to about
- 15 1,200 cP);
- 16 ii) having a Peel Adhesion Failure Temperature ("PAFT") of
- 17 greater than or equal to 32°C (90°F);
- 18 iii) having a Shear Adhesion Failure Temperature ("SAFT") of
- 19 greater than or equal to 93°C.(200°F) ; and
- 20 iv) exhibits 100% paper tear at 60°C (140°F).
- 21
- 22 7. The hot melt adhesive composition of any of Claims 2,3,4, or 6 wherein;
- 23 A) the homogeneous ethylene/ α -olefin interpolymers is an interpolymers of
- 24 ethylene and one or more C₃-C₃₀ α -olefins; and
- 25 B) the one or more tackifiers is selected from the group consisting of
- 26 aliphatic hydrocarbon resins, hydrogenated hydrocarbon resins, C₅ aliphatic or aromatic
- 27 hydrocarbon resins or an aromatically modified C₅ aliphatic or aromatic hydrocarbon
- 28 resins and combinations thereof; and
- 29 wherein the adhesive composition is capable of binding a fibrous cellulosic article to an
- 30 article selected from the group consisting of a fibrous cellulosic article, wood, metal,
- 31 glass, plastic, and combinations thereof..
- 32

- 1 8. The hot melt adhesive composition of Claims 4 or 5 wherein;
2 A) in the homogeneous ethylene/ α -olefin interpolymers, the α -olefin is
3 selected from the group consisting of from C₈ (1-Octene); C₁₀ (1-Decene), C₁₂ (1-
4 dodecene), C₁₄ (1-duodecene), C₁₄ (1-tetradecene), C₁₆ (1-hexadecene), C₁₈ (1-
5 octadecene), C₂₀₋₂₄⁺, C₂₄₋₂₈ and C₃₀ and combinations thereof; and
6 B) the one or more tackifiers is characterized by having an acid number
7 between 0 to and about 25.8.
8
9 9. The hot melt adhesive composition of Claim 7 or 8, further comprising one or
10 more compounds chosen from the group consisting of stabilizers, plasticizers, fillers,
11 antioxidants, preservatives, synergists, dyes, and pigments.
12
13 10. The hot melt adhesive composition of Claim 8, wherein the α -olefin is selected
14 from the group consisting of 1-octene and propylene.
15
16 11. The hot melt adhesive composition of Claim 8 or 10, wherein the copolymer has
17 a molecular weight distribution (Mw/Mn) ranging from about 2.1 – about 16.
18
19 12. The hot melt adhesive composition of Claim 8, 10 or 11, wherein the tackifier is
20 present in an amount of from about 15 to about 35 percent by weight (based on the final
21 weight of the hot melt adhesive composition).
22

1 13. A cellulosic article formed using a hot melt adhesive composition, the adhesive
2 composition consisting essentially of:

3 A) from about 40 to 100 percent by weight (based on the final weight of
4 the hot melt adhesive composition) of a homogeneous ethylene/ α -olefin interpolymers,
5 the homogeneous ethylene/ α -olefin interpolymers being characterized by having a
6 number average molecular weight (M_n) of from about 1,000 to about 9,000; and

7 B) from 0 to about 60 percent by weight (based on the final weight of the
8 hot melt adhesive composition) of one or more tackifiers.

9

10 14. The cellulosic article of Claim 13 wherein in the hot melt adhesive composition:

11 A) the homogeneous ethylene/ α -olefin interpolymers is present in an
12 amount of from about 60 to about 85 percent by weight (based on the final weight
13 of the hot melt adhesive composition) and the homogeneous ethylene/ α -olefin
14 interpolymers is further characterized by having:

15 i) a density of from about 0.880 to about 0.930 g/cm³; and

16 ii) a Brookfield Viscosity (measured at 149°C) ((300°F)) of from
17 about 5 to about 70 grams/(cm.second) (about 500 to about 7,000 cP); and

18 B) the one or more tackifiers is present in an amount of from about 15 to
19 about 40 percent by weight (based on the final weight of the hot melt
20 adhesive composition); and wherein

21 C) the hot melt adhesive composition is characterized by having:

22 i) a Brookfield Viscosity (measured at 177°C) ((350°F)) of from
23 about 4 to about 20 grams/(cm.second) (about 400 to about 2,000 cP);

24 ii) a Peel Adhesion Failure Temperature ("PAFT") of greater than
25 or equal to 43°C (110°F); and

26 iii) a Shear Adhesion Failure Temperature ("SAFT") of greater
27 than or equal to 60°C (140°F).

28

29 15. The cellulosic article of Claim 13, wherein in the hot melt adhesive composition;

30 A) the homogeneous ethylene/ α -olefin interpolymers is present in an
31 amount of from about 65 to about 80 percent by weight (based on the final weight of the

- 1 hot melt adhesive composition) and the homogeneous ethylene/ α -olefin interpolymers is
2 further characterized by having:
- 3 i) a density of from about 0.890 to about 0.920 g/cm³;
 - 4 ii) a number average molecular weight (M_n) of from about 1,250 to
5 about 7,000; and
 - 6 iii) a Brookfield Viscosity (measured at 149°C) ((300°F)) of from
7 about 10 to about 60 grams/(cm.second) (about 1,000 to about
8 6,000 cP); and
- 9 B) the one or more tackifiers is present in an amount of from about 20 to
10 about 35 percent by weight (based on the final weight of the hot melt
11 adhesive composition); and wherein
- 12
- 13 C) the hot melt adhesive composition is characterized by:
- 14 i) having a Brookfield Viscosity (measured at 177°C) ((350°F)) of
15 from about 5 to about 14 grams/(cm.second) (about 500 to about
16 1,400 cP);
 - 17 ii) having a Peel Adhesion Failure Temperature ("PAFT") of
18 greater than or equal to 46°C (115°F);
 - 19 iii) having a Shear Adhesion Failure Temperature ("SAFT") of
20 greater than or equal to 66°C (150°F); and
 - 21 iv) exhibiting 100% paper tear at 60°C (140°F).
- 22

23 16. The cellulosic article of Claim 13 wherein in the hot melt adhesive composition:

- 24 A) the homogeneous ethylene/ α -olefin interpolymers is further
25 characterized by having:
- 26 i) a density of from about 0.893 to about 0.930 g/cm³;
 - 27 ii) a number average molecular weight (M_n) of from about 1,000
28 to about 6,000; and
 - 29 iii) a Brookfield Viscosity (measured at 149°C) ((300°F)) of from
30 about 15 to about 50 grams/(cm.second) (about 1,500 to about
31 5,000 cP); and
- 32 B) the hot melt adhesive composition is characterized by:

- 1 i) having a Brookfield Viscosity (measured at 177°C) ((350°F)) of
2 from about 4 to about 14 grams/(cm.second) (about 400 to about
3 1,400 cP) ;
4 ii) having a Peel Adhesion Failure Temperature ("PAFT") of
5 greater than or equal to 32°C (90°F);
6 iii) having a Shear Adhesion Failure Temperature ("SAFT") of
7 greater than or equal to 93°C. (200°F.); and
8 iv) exhibits 100% paper tear at 49°C (120°F).
9

10 17. The cellulosic article of Claim 16 wherein in the hot melt adhesive composition;

11 A) the homogeneous ethylene/ α -olefin interpolymer is characterized by
12 having:

- 13 i) a density of from about 0.894 to about 0.910 g/cm³;
14 ii) a number average molecular weight (Mn) of from about 1,100
15 to about 5,300; and
16 iii) a Brookfield Viscosity (measured at 149°C)((300°F)) of from
17 about 16 to about 32 grams/(cm.second) (about 1,600 to about
18 3,200 cP); and

19 B) the hot melt adhesive composition is characterized by:

- 20 i) having a Brookfield Viscosity (measured at 177°C) ((350°F)) of
21 from about 7 to about 12 grams/(cm.second) (about 700 to about
22 1,200 cP);
23 ii) having a Peel Adhesion Failure Temperature ("PAFT") of
24 greater than or equal to 32°C (90°F);
25 iii) having a Shear Adhesion Failure Temperature ("SAFT") of
26 greater than or equal to 93°C.(200°F) ; and
27 iv) exhibits 100% paper tear at 60°C (140°F).
28

29 18. The cellulosic article of Claims 16 or 17, wherein in the hot melt adhesive
30 composition:

1 A) the homogeneous ethylene/ α -olefin interpolymer is an interpolymer of
2 ethylene and one or more C_3 - C_{30} α -olefins; and

3 B) the one or more tackifiers is selected from the group consisting of aliphatic
4 hydrocarbon resins, hydrogenated hydrocarbon resins, C_5 aliphatic or aromatic
5 hydrocarbon resins or an aromatically modified C_5 aliphatic or aromatic hydrocarbon
6 resins and combinations thereof.

7
8 19. The cellulosic article of Claim 18, wherein in the hot melt adhesive composition;

9 A) in the homogeneous ethylene/ α -olefin interpolymer, the α -olefin is
10 selected from the group consisting of from C_8 (1-Octene); C_{10} (1-Decene), C_{12} (1-
11 dodecene), C_{14} (1-duodecene), C_{14} (1-tetradecene), C_{16} (1-hexadecene), C_{18} (1-
12 octadecene), C_{20-24+} , C_{24-28} and C_{30} and combinations thereof; and

13 B) the one or more tackifiers is characterized by having an acid number
14 between 0 to and about 25.8.

15

16 20. The cellulosic article of Claims 19, wherein the cellulosic article is selected from
17 the group consisting of corrugated cardboard, kraft paper, linerboard, and paper.

18

19 21. A method of producing a polymer composition, the method comprising the steps of:
20 admixing:

21 an amount of from about 60 to about 85 percent by weight (based on the final
22 weight of the polymer composition) of a homogeneous ethylene/ α -olefin interpolymer;
23 the homogeneous ethylene/ α -olefin interpolymer characterized by having:

24 i) a density of from about 0.880 to about 0.930 g/cm³;

25 ii) a number average molecular weight (Mn) of from about 1,000
26 to about 9,000; and

27 iii) a Brookfield Viscosity (measured at 149°C) (300°F) of from
28 about 5 to about 70 grams/(cm.second) (about 500 to about 7,000 cP); and

29

30 admixing an amount of from 15 to about 40 percent by weight (based on the final
31 weight of the polymer composition) of a tackifier; and wherein

32 the polymer composition is characterized by having:

- 1 i) a Brookfield Viscosity (measured at 177°C) (350°F) of from
2 about 4 to about 20 grams/(cm.second) (about 400 to about 2,000 cP);
3 ii) a Peel Adhesion Failure Temperature ("PAFT") of greater than
4 or equal to 43°C (110°F); and
5 iii) a Shear Adhesion Failure Temperature ("SAFT") of greater
6 than or equal to 60°C (140°F).

7

8 22. The method as described in claim 21, wherein:

9 A) the homogeneous ethylene/ α -olefin interpolymer is characterized by
10 having:

- 11 i) a density of from about 0.893 to about 0.930 g/cm³;
12 ii) a number average molecular weight (Mn) of from about 1,000
13 to about 6,000; and
14 iii) a Brookfield Viscosity (measured at 149°C) (300°F) of from
15 about 15 to about 50 grams/(cm.second) (about 1,500 to about
16 5,000 cP); and

17 B) the polymer composition is characterized by:

- 18 i) having a Brookfield Viscosity (measured at 177°C) (350°F) of
19 from about 4 to about 14 grams/(cm.second) (about 400 to about
20 1,400 cP);
21 ii) having a Peel Adhesion Failure Temperature ("PAFT") of
22 greater than or equal to 32°C (90°F);
23 iii) having a Shear Adhesion Failure Temperature ("SAFT") of
24 greater than or equal to 93°C. (200°F.) ; and
25 iv) exhibits 100% paper tear at 60°C (140°F).

26